

Review

The Next Frontier in Energy Storage: A Game-Changing Guide to Advances in Solid-State Battery Cathodes

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Abstract: As global energy priorities shift toward sustainable alternatives, the need for innovative energy storage solutions becomes increasingly crucial. In this landscape, solid-state batteries (SSBs) emerge as a leading contender, offering a significant upgrade over conventional lithium-ion batteries in terms of energy density, safety, and lifespan. This review provides a thorough exploration of SSBs, with a focus on both traditional and emerging cathode materials like lithium cobalt oxide (LiCoO_2), lithium manganese oxide (LiMn_2O_4), lithium iron phosphate (LiFePO_4), as well as novel sulfides and oxides. The compatibility of these materials with solid electrolytes and their respective benefits and limitations are extensively discussed. The review delves into the structural optimization of cathode materials, covering strategies such as nanostructuring, surface coatings, and composite formulations. These are critical in addressing issues like conductivity limitations and structural vulnerabilities. We also scrutinize the essential roles of electrical and thermal properties in maintaining battery safety and performance. To conclude, our analysis highlights the revolutionary role of SSBs in the future of energy storage. While substantial advancements have been made, the path forward presents numerous challenges and research opportunities. This review not only acknowledges these challenges, but also points out the need for scalable manufacturing approaches and a deeper understanding of electrode–electrolyte interactions. It aims to steer the scientific community toward addressing these challenges and advancing the field of SSBs, thereby contributing significantly to the development of environmentally friendly energy solutions.



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1. Introduction

The global transition from fossil fuels to cleaner and more sustainable energy sources is an imperative response to the growing environmental concerns associated with the use of conventional hydrocarbon-based fuels [1]. The detrimental impacts of greenhouse gas emissions, climate change, and resource depletion have accelerated the need for innovative solutions to reduce our reliance on fossil fuels and mitigate their environmental footprint [1,2]. In this era of sustainable energy, high-performance energy storage systems play an important role in ensuring the reliability and efficiency of renewable energy sources [3,4]. These systems bridge the gap between energy generation and consumption, enabling the effective utilization of intermittent sources like wind and solar power while enhancing grid stability and resilience [3,4].

In the landscape of energy storage, solid-state batteries (SSBs) are increasingly recognized as a transformative alternative to traditional liquid electrolyte-based lithium-ion batteries, promising unprecedented advancements in energy density, safety, and longevity [5–7]. These benefits stem from the incorporation of advanced electrode materials and solid-state electrolytes, which together enable heightened energy storage capacities. Notably, the

absence of flammable liquid electrolytes in SSBs mitigates the risk of thermal runaway, a paramount safety concern, especially in applications like electric vehicles (EVs) and portable electronics [8–11]. Beyond safety, SSBs, with their augmented energy densities, champion the development of more compact, energy-efficient devices [11–16]. Their resilience against dendrite formation and extended cycle life further accentuates their suitability for applications demanding sustained reliability and long-term energy storage [15–18].

However, despite the remarkable promise of SSBs, their commercialization remains constrained by specific challenges, with the cathode being a pivotal component significantly affecting battery performance [19,20]. This comprehensive review aims to synthesize the state-of-the-art advancements in solid-state battery cathodes, shedding light on both material chemistry and engineering techniques that contribute to enhanced performance metrics. Through an in-depth exploration of cathode materials, structural optimization, electrical properties, thermal characteristics, and practical considerations, this review provides a holistic perspective on the development of SSB cathodes. The following sections will explore traditional cathode materials and emerging alternatives, discuss strategies for structural optimization, address practical integration aspects, conduct a comparative evaluation of cathode materials, and finally, identify existing research gaps and propose future directions in the development of SSB cathodes.

The topics that will be discussed (challenges in solid-state battery development, traditional cathode materials, emerging cathode materials, the structural optimization of cathode materials, the integration of advanced cathode materials into SSBs, a comparative evaluation of cathode materials, the identification of research gaps, and challenges in future directions in SSBs cathode development) are presented in Figure 1.

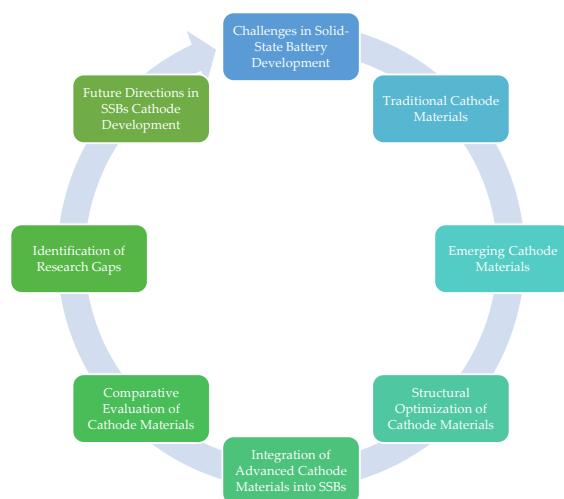


Figure 1. Schematic of the topics discussed in the review.

Challenges in Solid-State Battery Development

Solid-state batteries (SSBs) represent a promising advancement in energy storage technology, offering higher energy density and improved safety compared to conventional lithium-ion batteries. However, several challenges impede their widespread adoption. A critical issue is the interface instability between solid electrolytes and electrodes [11]. Solid electrolytes, while advantageous for their non-flammability and potentially higher ionic conductivity, often suffer from poor interface contact with electrodes. This poor contact can lead to increased resistance and decreased ion transport efficiency, significantly impacting the battery's overall performance and lifespan [2,10]. Additionally, the formation of interphase layers at these interfaces can further exacerbate resistance issues. Another major challenge is related to lithium dendrite growth during cycling in SSBs with lithium metal anodes. The growth of these dendrites can penetrate solid electrolytes, leading to short-circuiting and potential safety hazards [15,18].

The mechanical properties of solid electrolytes present another significant hurdle. Many solid electrolytes are brittle and may crack under mechanical stress, which is particularly problematic given the volume changes that electrodes undergo during charge and discharge cycles [4,7]. This brittleness also complicates the cell manufacturing process, as it requires delicate handling and precise engineering to ensure structural integrity. Additionally, achieving high ionic conductivity in solid electrolytes, comparable to liquid electrolytes, remains a challenge. Although some solid electrolytes have demonstrated high ionic conductivities at room temperature, many still lag behind their liquid counterparts, especially under various temperature conditions [9,12].

Another critical issue is thermal management. Unlike traditional lithium-ion batteries where liquid electrolytes can help dissipate heat, the solid electrolytes in SSBs often have lower thermal conductivities, potentially leading to localized hotspots and uneven temperature distribution within the cell [6,16]. This uneven thermal profile can exacerbate the degradation of both the electrolyte and electrode materials, further hindering the battery's performance and cycle life. Moreover, the integration of high-capacity materials like sulfur or silicon, which are seen as potential candidates for increasing the energy density of SSBs, introduces additional complexities. These materials undergo significant volume changes during cycling, which, combined with the brittle nature of solid electrolytes, raises concerns about the mechanical integrity and long-term stability of the cell [7,13]. Consequently, addressing these thermal and mechanical challenges is essential for advancing the development of SSBs, particularly for applications demanding high energy density and safety, such as in electric vehicles and aerospace technologies.

2. Traditional Cathode Materials

Lithium cobalt oxide (LiCoO_2) has been a foundation in the development of lithium-ion batteries. It offers high energy density but comes with challenges such as safety risks and limited thermal stability [21]. The original ideas for LiCoO_2 as a cathode material were inspired by interdisciplinary research in solid-state physics and chemical structure bonding [21]. A study by Wei et al. [22] demonstrated that porous LiCoO_2 fabricated from metal–organic frameworks (see Figure 2) show excellent stability and superior rate capability, delivering a reversible capacity of 106.5 mAh g^{-1} at 2C with a stable capacity retention of 96.4% even after 100 cycles.

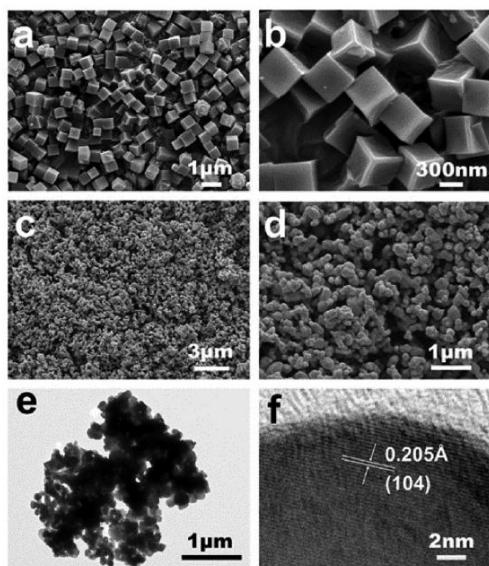


Figure 2. Scanning electron microscope (SEM) images of (a,b) ZIF-67 (Co) and (c,d) porous LiCoO_2 at varying magnifications; (e) transmission electron microscope (TEM) image and (f) high-resolution transmission electron microscope (HR-TEM) image of porous LiCoO_2 (reprinted with permission from ref. [22], Copyright 2020, Royal Society of Chemistry).

LiCoO_2 has a theoretical capacity of 274 mAh g^{-1} but often fails to deliver more than half of this due to structural deformation [23]. The material's high energy density makes it suitable for applications requiring compact and lightweight batteries, such as mobile devices and certain types of electric vehicles. A recent study by Cherkashinin et al. [24] investigated into the intrinsic stability of LiCoO_2 and found that the material exhibits fully reversible electronic properties after the first electrochemical cycle, providing insights into the development of doping strategies to enhance its electronic conductivity.

Lithium manganese oxide (LiMn_2O_4) is considered an environmentally friendly and cost-effective alternative to LiCoO_2 . It has a theoretical capacity of 148 mAh g^{-1} and can deliver more than 95% of its theoretical capacity [23]. However, it suffers from low conductivity, manganese dissolution in the electrolyte, and structural distortion at elevated temperatures [23]. Various strategies have been employed to improve LiMn_2O_4 's performance, including structure doping with single or multiple cations and anions, and surface modification by coating with materials like carbonaceous compounds, oxides, and phosphates [25]. Doping stabilizes the LiMn_2O_4 spinel structure and reduces the amount of electrochemically active Mn^{3+} , which is responsible for manganese dissolution into the electrolyte [26]. A study by Choi et al. [27] compared the performance of LiMn_2O_4 , with nickel–cobalt manganese (LiNiMnCoO_2), and lithium–iron phosphate (LiFePO_4), and found that it exhibits good thermal stability and capacity retention under various driving cycles.

Lithium iron phosphate (LiFePO_4) is another alternative cathode material known for its robustness and safety [28]. It has a theoretical capacity of 170 mAh g^{-1} and is particularly stable during charge and discharge cycles [28]. LiFePO_4 is often used in applications where safety and long cycle life are more critical than energy density, such as in large-scale energy storage systems and certain electric vehicles. In a study focusing on the temperature's effect on different cathode materials, LiFePO_4 was found to have optimal performance in a temperature range of $20\text{--}50^\circ\text{C}$ [29]. The study also highlighted that the state of charge (SOC) has a significant impact on the internal resistance of the battery, affecting its overall performance [30]. Dong et al. [31] developed an adhesive bonding solid polymer electrolyte (ABSPE) for zinc-ion batteries and demonstrated enhanced anti-aging properties, maintaining constant surface resistance for over 200 h, and achieved an ionic conductivity of $3.77 \times 10^{-4} \text{ S cm}^{-1}$. This ABSPE exhibited a diffusion-controlled mechanism in a $\text{Zn/ABSPE}/\beta\text{-MnO}_2$ system, unlike the capacitive dominated process in hydrogel electrolytes, and enabled the fabrication of a flexible device with superior power performance under various physical stresses (see Figure 3).

The interaction between cathode materials, solid electrolytes, particle size, and binder type plays a pivotal role in dictating the performance, safety, and longevity of solid-state batteries. Understanding and optimizing these correlations for LiCoO_2 , LiMn_2O_4 , and LiFePO_4 cathodes are essential for the advancement of solid-state battery technologies. The correlation between these properties is presented below:

LiCoO_2 (lithium cobalt oxide): LiCoO_2 is known for its high energy density, but its performance is closely tied to the choice of solid electrolyte and the microstructural characteristics of the cathode. The optimal electrolyte for LiCoO_2 in solid-state batteries should have high ionic conductivity and compatibility with the cathode material to prevent interface degradation. For instance, solid electrolytes like lithium lanthanum zirconate (LLZO) demonstrate good compatibility with LiCoO_2 due to their chemical stability. Regarding particle size, smaller LiCoO_2 particles have a larger surface area, which can enhance electrode–electrolyte contact but may also lead to more significant structural instability during cycling. Binder choice in LiCoO_2 cathodes is also critical; polyvinylidene fluoride (PVDF) is commonly used due to its good adhesion properties and chemical stability, ensuring the integrity of the cathode structure during charge–discharge cycles.

LiMn_2O_4 (lithium manganese oxide): The performance of LiMn_2O_4 is significantly influenced by its interaction with the electrolyte, particularly concerning manganese dissolution issues. Solid electrolytes that provide a stable interface and minimize the dissolution

of manganese, such as lithium phosphorus oxynitride (LiPON), are preferred for enhancing longevity and safety. Particle size plays a crucial role in LiMn_2O_4 cathodes, with nano-sized particles offering an improved rate capability and a reduced path length for lithium-ion diffusion. However, nano-sizing can exacerbate manganese dissolution, necessitating careful optimization. The choice of binder in LiMn_2O_4 cathodes, such as carboxymethyl cellulose (CMC), is vital for maintaining electrode integrity, especially considering the material's susceptibility to volume changes during cycling.

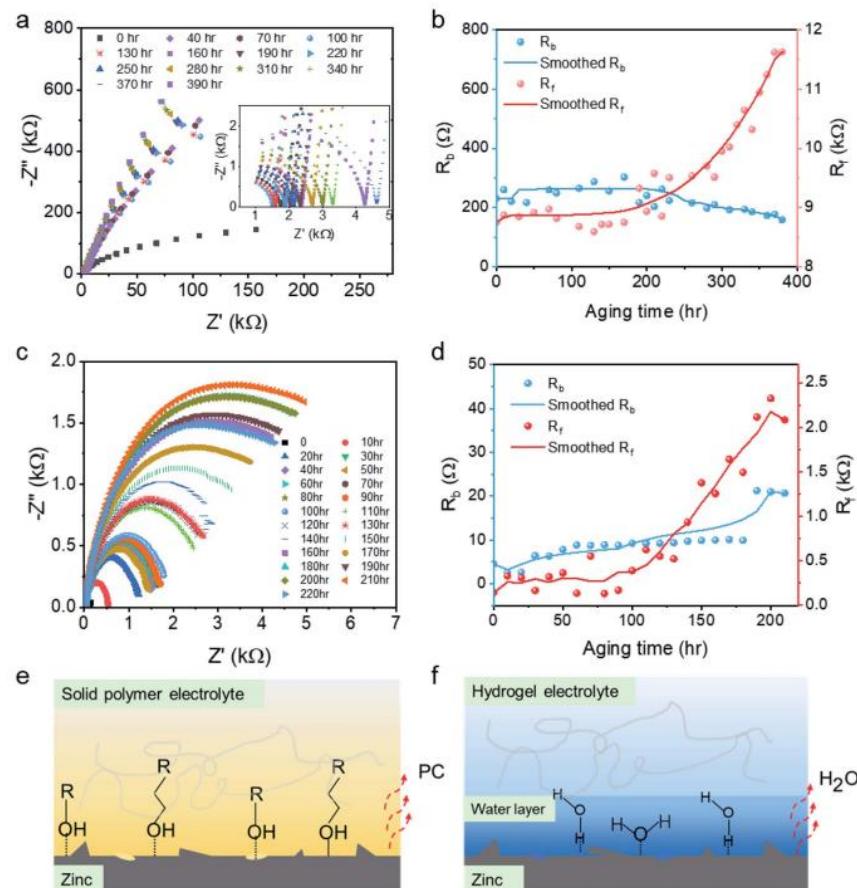


Figure 3. (a) Electrochemical impedance spectroscopy (EIS) aging test of adhesive bonding solid polymer electrolyte (Zn/ABSPE/Zn); (b) analysis of bulk and surface resistances in ABSPE; (c) EIS aging assessment for alginate hydrogel polymer electrolyte (Zn/HPE/Zn); (d) evaluation of bulk and surface resistances in alginate hydrogel polymer electrolyte; (e) assessment of surface intermolecular forces in ABSPEs; (f) analysis of surface intermolecular forces in HPEs (reprinted with permission from ref. [31], Copyright 2020, *Journal of Materials Chemistry A*).

LiFePO₄ (lithium iron phosphate): LiFePO₄'s stability and safety are enhanced when paired with solid electrolytes that exhibit thermal stability and robustness, such as garnet-type electrolytes like LLZO. The particle size of LiFePO₄ directly impacts its rate performance; smaller particles facilitate faster lithium-ion diffusion but can lead to increased side reactions at the electrode–electrolyte interface. Binders that can accommodate the volume expansion of LiFePO₄ while maintaining good electrode integrity are crucial. Binders like poly(acrylic acid) (PAA) have been found to be effective, offering strong mechanical adhesion and contributing to the overall structural stability of the cathode.

Importance of Solid Electrolytes in Solid-State Batteries

The crucial role of SSBs extends far beyond the mere replacement of liquid counterparts. Solid electrolytes stand at the forefront of revolutionizing energy storage systems,

primarily due to their intrinsic safety features and ability to enable high energy densities [12]. The non-flammable nature of solid electrolytes, typically composed of inorganic ceramics or polymers, eliminates the risk of leakage and thermal runaway, a significant concern in conventional lithium-ion batteries with organic liquid electrolytes [12,20,24]. This critical safety enhancement makes SSBs particularly suitable for high-energy-demand applications, including electric vehicles and portable electronics, where thermal stability is paramount [26,29].

Solid electrolytes also facilitate the integration of lithium metal anodes, as opposed to the traditional carbonaceous anodes, thus unlocking the potential for higher energy densities. Lithium metal anodes, in synergy with solid electrolytes, could theoretically provide an energy density nearly double that of current lithium-ion batteries [16]. This is chiefly due to the higher specific capacity of lithium metal (3860 mAh g^{-1}) compared to graphite (372 mAh g^{-1}). However, the compatibility between the solid electrolyte and the lithium metal anode remains a challenging area, particularly in terms of maintaining a stable interface and suppressing lithium dendrite growth during electrochemical cycling [20,26,29]. Moreover, solid electrolytes present an opportunity to re-envision battery architectures. The rigidity and stability of solid electrolytes allow for thinner and more flexible cell designs, potentially leading to more compact and versatile battery configurations. This advantage is particularly significant for wearable electronics and implantable medical devices, where form factor plays a crucial role [14,16].

Despite these advantages, the development of solid electrolytes with high ionic conductivity comparable to liquid electrolytes remains a substantial challenge. The pursuit of solid electrolytes with high lithium-ion transference numbers, minimal electronic conductivity, and adequate mechanical properties is a key focus area in current research. Recent advancements have highlighted the promise of garnet-type electrolytes, like $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO), which exhibit high ionic conductivity and stability against lithium metal [20,28]. However, addressing issues such as interfacial resistance, especially at lower operating temperatures, and ensuring long-term chemical and electrochemical stability are critical for the practical realization of high-performance SSBs. These material and interface challenges underscore the need for ongoing research in solid electrolyte development, targeting a balance between conductivity, stability, and mechanical properties to meet the demanding requirements of next-generation energy storage systems.

3. Emerging Cathode Materials

Sulfide-based cathode materials have gained significant attention in the realm of all-solid-state lithium batteries (ASSLBs) [32–36]. These materials offer promising attributes, especially when paired with solid-state sulfide electrolytes (SSSEs). A recent study [34] introduced a novel $\text{Li}_{2.96}\text{P}_{0.98}\text{S}_{3.92}\text{O}_{0.06}$ - Li_3N glass–ceramic electrolyte (GCE) where O and N substitution produced unique functional units that enabled superior ionic conductivity at room temperature. Notably, these units in the $\text{Li}_{2.96}\text{P}_{0.98}\text{S}_{3.92}\text{O}_{0.06}$ - Li_3N effectively prevent structural degradation against moisture. Furthermore, this GCE addresses key challenges and showcases potential for use in high-energy ASSLBs.

Emerging oxide cathodes in the context of ASSLBs refer to newer compositions or structural modifications of oxide materials that potentially offer improved energy densities, safety profiles, and compatibility with solid electrolytes. These include novel formulations or structurally engineered oxides that are different from the traditional materials in their electrochemical behavior, safety profiles, or other key characteristics.

Oxide-based cathode materials, when paired with solid electrolytes, can significantly enhance the energy density of ASSLBs. The transition from liquid-based Li-ion batteries (LIBs) to ASSLBs has been driven by the potential advantages of oxide- and sulfide-based solid electrolytes [35]. A strategic approach to this transition involves analyzing the pairing of solid electrolytes with oxide cathode materials and the Li metal anode [35]. The chemical, electrochemical, and mechanical properties of these solid electrolytes play a crucial role in determining the performance of the resultant ASSLBs [35]. Ban et al. [35] developed

a composite solid electrolyte (CSE) membrane composed of poly(ethylene oxide) (PEO), LiClO₄, and Li 1.3 Al 0.3 Ti 1.7(PO₄)₃ (LATP) using a solution casting technique. Their findings indicated that the membrane exhibited high lithium-ion conductivity (approximately 1×10^{-3} S cm⁻¹) alongside superior mechanical and electrochemical properties. Notably, the solid-state battery utilizing this membrane demonstrated a capacity of 109.3 mAh g⁻¹ at a 1C rate after 500 cycles, suggesting an effective approach for designing high-performance solid-state batteries, as depicted in Figure 4.

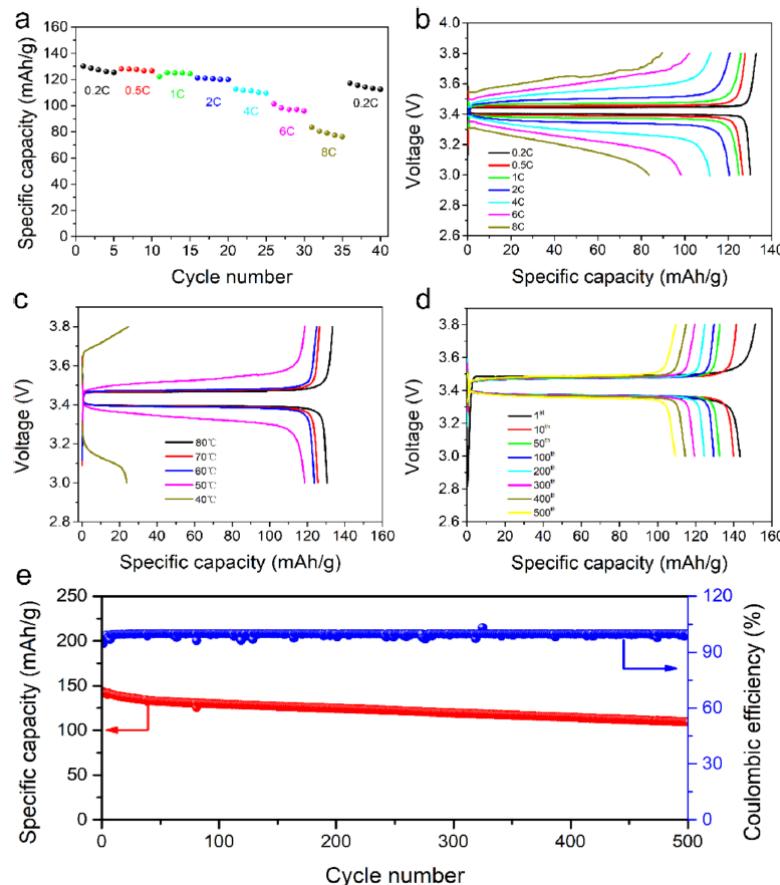


Figure 4. Scanning electron microscope (SEM) images of (a,b) ZIF-67 (Co) and (c,d) porous LiCoO₂ at varying magnifications; (e) transmission electron microscope (TEM) image and high-resolution transmission electron microscope (HR-TEM) image of porous LiCoO₂ (reprinted with permission from ref. [35], Copyright 2018, American Chemical Society).

Emerging cathode materials present a myriad of advantages and challenges that influence their potential in next-generation energy storage systems. While they offer benefits such as fuel flexibility and environmental friendliness, they also come with inherent limitations that need to be addressed for optimal performance. Table 1 presents some of the advantages and limitations of emerging cathode materials.

Table 1. Advantages and limitations of emerging cathode materials.

Category	Aspect	Description and Performance Metrics	Reference
Advantages			
	High Energy Density	Emerging cathode materials in SSBs, like lithium nickel manganese cobalt oxide (NMC), offer higher energy density (up to 250–300 Wh/kg) compared to traditional lithium-ion batteries. This makes them suitable for longer-range electric vehicles.	[32]
	Enhanced Safety	Solid electrolytes in SSBs significantly reduce the risk of leakage and combustion. They are non-flammable and have a higher thermal stability, making them safer under extreme conditions.	[33]
	Longer Lifespan	Advanced cathode materials in SSBs show reduced degradation, with potential cycle lives exceeding 5000 cycles, significantly higher than conventional lithium-ion batteries, which typically last for about 1000–2000 cycles.	[34]
Limitations			
	High Manufacturing Cost	The production of SSBs is more expensive due to complex manufacturing processes and the use of novel, sometimes scarce, materials. This cost is a significant barrier to widespread adoption and commercialization.	[35]
	Limited Power Density	Despite high energy density, SSBs currently have lower power density (around 1000–2000 W/kg) compared to traditional lithium-ion batteries. This limitation affects their ability to quickly charge or provide high power bursts.	[35]
	Challenges in Scalability	Scaling up production to meet large-scale demand is challenging due to the intricate manufacturing processes and the need for high precision and quality control. This issue is a key focus for research and development in the field.	[36]
	Operational Temperature	SSBs often have a narrower operational temperature range compared to liquid electrolyte batteries. This can limit their use in extreme weather conditions, though ongoing research aims to improve this aspect.	[37]

The compatibility of cathode materials with solid electrolytes is a critical factor in the performance of ASSLBs. For instance, lithium aluminum germanium phosphate (LAGP) is a solid electrolyte that has gained significant attention due to its stability in air and good ionic conductivity [37]. However, challenges like poor interface compatibility with Li anodes and slow Li-ion conduction in thick pellets have been identified [37]. Advanced interface engineering strategies, such as introducing a functional interlayer, have been proposed to address these challenges [37].

As a summary of this section, emerging cathode materials in ASSLBs include sulfide-based and oxide-based types. Sulfide-based materials, especially when used with solid-state sulfide electrolytes (SSSEs), offer promising attributes like enhanced ionic conductivity and structural stability against moisture, as shown in the $\text{Li}_{2.96}\text{P}_{0.98}\text{S}_{3.92}\text{O}_{0.06}\text{-Li}_3\text{N}$ GCE study. Oxide cathodes represent a shift in composition and structure from traditional materials, aiming for higher energy densities and safety. Their pairing with solid electrolytes like poly(ethylene oxide) (PEO) and $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ (LATP) in CSE membranes results in batteries with impressive performance, as evidenced by Ban et al.'s study. However, limitations such as interface compatibility issues and ion conduction challenges in thick pellets persist. Lithium aluminum germanium phosphate (LAGP) is a notable solid electrolyte with good stability and conductivity but suffers from poor Li-anode compatibility and slow Li-ion conduction, highlighting the need for advanced interface engineering.

Future research directions in emerging cathode materials for ASSLBs should be focused on the following aspects. (a) Interface engineering for cathodes: Concentrating on

improving the interface compatibility between emerging cathode materials and solid-state electrolytes. This involves developing new materials or surface treatments that enhance interfacial contact, reduce resistance, and improve ion transport. (b) Structural optimization of cathode materials: Exploring advanced structural engineering of cathode materials to optimize their ionic and electronic conductivity. This includes investigating different crystal structures, doping strategies, and nanostructuring to enhance performance. (c) Development of composite cathode materials: Investigating composite cathode materials that integrate various functional components. This research would aim at combining different materials to exploit synergistic effects, thereby improving energy density, conductivity, and stability. (d) Targeting cathode-specific limitations: Addressing challenges specific to emerging cathode materials, such as sensitivity to moisture, mechanical stability, and thermal performance. Research should focus on enhancing the intrinsic properties of these cathodes to overcome these limitations. (e) Assessing environmental and safety aspects: Evaluating the environmental impact and safety profiles of new cathode materials. This includes studying the toxicity, recyclability, and life cycle analysis of these materials to ensure sustainable and safe deployment. (f) Enhancing cathode stability and longevity: Focusing on the long-term stability and durability of emerging cathode materials. Research should aim to understand and mitigate degradation mechanisms to extend the life and performance of ASSLBs. (g) Scalable synthesis and manufacturing: Developing scalable and cost-effective synthesis and manufacturing processes for emerging cathode materials. This includes the innovation of production techniques that are suitable for large-scale applications, maintaining quality and performance consistency.

4. Structural Optimization of Cathode Materials

4.1. Nanostructuring

The structural optimization of cathode materials in SSBs plays a crucial role in enhancing their electrochemical performance. Nanostructuring is a promising approach that involves reducing the dimensions of cathode materials to the nanoscale [38]. This strategy offers several advantages in terms of improved conductivity and enhanced surface area for electrochemical reactions [38–40].

The nanostructuring of cathode materials has been demonstrated in various studies. For instance, Sun et al. [38] demonstrated the advantages of utilizing a Li₂WO₄-coated LiCoO₂ cathode in combination with a sulfide Li₆PS₅Cl solid electrolyte, enhancing the electrochemical performance of all-solid-state batteries. In a similar vein, Liu et al. [40] enhanced the interface between the superionic conductor and the polymer electrolyte (polyvinylidene fluoride) through the in situ creation of a pyrochlore-type La₂Sn₂O₇ (LSO) ceramic layer on Li_{6.4}La₃Zr_{1.4}Ta_{0.6}O₁₂ (LLZTO). They noted that the LSO synthesis process depletes La in LLZTO, leading to an increased concentration of Li ions in LLZTO and substantially boosting the conductivity of the composite electrolyte (LLZTO@LSO-CSE). The conductivity of LLZTO@0.9%LSO-CSE showed a significant improvement, reaching as high as 1.30×10^{-41} S cm⁻¹, compared to 3.15×10^{-53} S cm⁻¹ for the unmodified sample. Moreover, after 400 cycles, the LiFePO₄ | LLZTO@0.9%LSO-CSE | Li battery maintained a discharge capacity of 110.6 mAh g⁻¹, a Coulombic efficiency of 99.4%, and a capacity retention rate of 72%, which is superior to the 75.5 mAh g⁻¹, 98% efficiency, and 49% retention rate of the pristine sample, as illustrated in Figure 5.

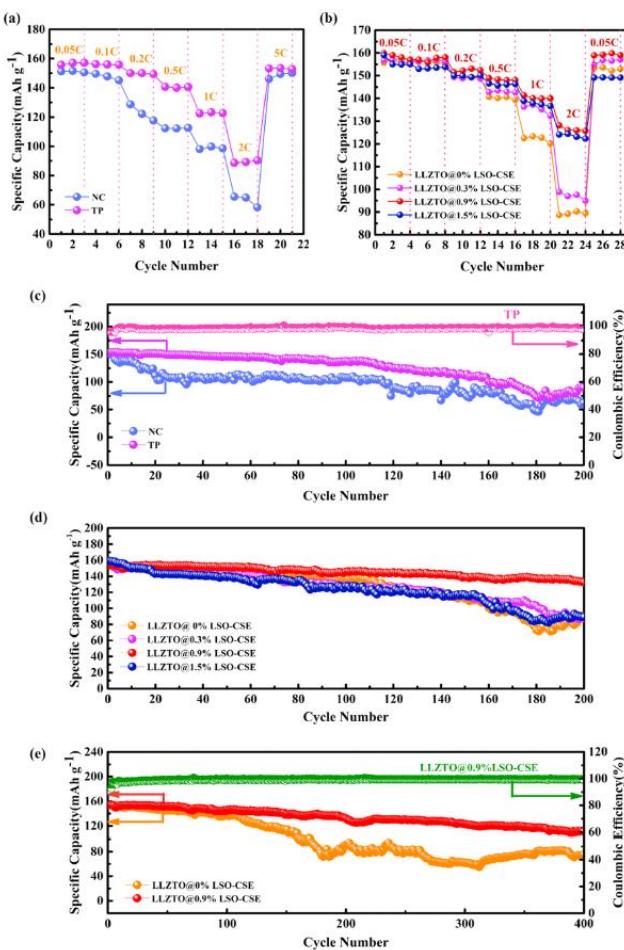


Figure 5. Performance evaluation of electrochemical aspects in the solid–state battery. **(a)** Capability analysis under different rates and **(c)** assessment of cycling stability at 0.2 C for TP and standard contact. **(b)** Examination of rate capabilities and **(d,e)** evaluation of cycling behavior at 0.2 C in solid-state batteries utilizing LLZTO@LSO-CSEs (reprinted with permission from ref. [40], Copyright 2021, American Chemical Society).

4.2. Surface Coatings

Surface coatings represent another avenue for improving cathode materials in SSBs. Coating cathode particles with conductive and protective materials can mitigate issues related to structural instability, reactivity with electrolytes, and enhance overall electrode performance [41,42]. Recent research by Liang et al. [43] focused on surface coating strategies for $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC) cathode materials. They demonstrated that a gradient oxy-thiophosphate coating improved the structural stability of NMC cathodes, reducing capacity fading and improving cycling stability. Additionally, surface coatings can enhance the safety of SSBs by preventing the formation of harmful solid electrolyte interphase (SEI) layers [44,45].

4.3. Composite Approaches

Composite cathode materials are designed by integrating various components to leverage their complementary properties. This approach combines the advantages of multiple materials to address the challenges associated with low conductivity and structural instability in SSB cathodes [46,47]. One notable example is the development of composite cathodes using conductive polymers. Researchers, such as Liu et al. [40], have successfully incorporated various strategies to improve the interface of cathode materials in solid-state

lithium batteries. The resulting approaches exhibited improved electronic conductivity and ion diffusivity, leading to enhanced rate capability and cycling stability.

Composite approaches also extend to incorporating ionic conductors within the cathode matrix. Liang et al. [44] observed that high-energy Ni-rich layered oxide cathode materials like $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ (NMC811) experience adverse side reactions and interfacial structural instability when paired with sulfide solid-state electrolytes in all-solid-state lithium-based batteries. To address this, their team introduced a gradient coating strategy for NMC811 particles using lithium oxy-thiophosphate ($\text{Li}_3\text{P}_{1+x}\text{O}_4\text{S}_{4x}$), achieved through the atomic layer deposition of Li_3PO_4 followed by an in situ development of a gradient $\text{Li}_3\text{P}_{1+x}\text{O}_4\text{S}_{4x}$ coating. This customized surface engineering of NMC811 prevents the structural degradation commonly seen in the transition from layered to spinel phases at grain boundaries and significantly enhances the stability of the cathode/solid electrolyte interface during cycling. In tests with an indium metal anode and a $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ solid electrolyte, the oxy-thiophosphate-coated NMC811 cathode demonstrated a specific discharge capacity of 128 mAh g^{-1} after nearly 250 cycles at 0.178 mA cm^{-2} and 25°C , as shown in Figure 6.

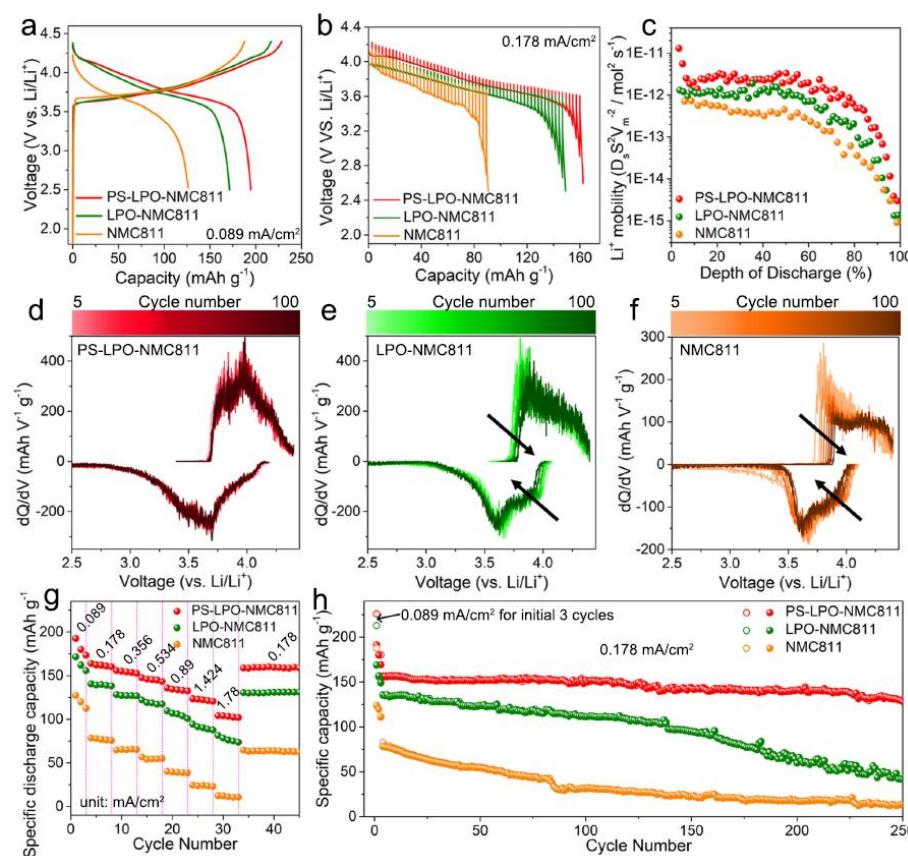


Figure 6. All-solid-state Li-ion-cell electrochemical performance with an In anode, $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ solid electrolyte, and various NCM811 cathodes at 25°C . (a) First-cycle charge/discharge profiles at 0.089 mA cm^{-2} . (b) Galvanostatic intermittent titration technique (GITT) curves recorded during the discharge phase, and (c) corresponding Li^+ diffusion coefficients for three types of NMC811 cathodes at various discharge states. (d–f) Differential capacity (dQ/dV) plots for the three NMC811 cathode variants over the first 100 cycles at 0.178 mA cm^{-2} . (g) Rate performance and (h) cycling stability of the three NMC811 cathode types. The PS-LPO-NMC811 cathode is shown in red, the LPO-NMC811 cathode in green, and the NMC811 cathode in orange (reprinted with permission from ref. [44], Copyright 2023, Springer Nature).

4.4. Addressing Issues Like Low Conductivity and Structural Instability

The structural optimization of SSB cathodes aims to mitigate two fundamental challenges: low electrical conductivity and structural instability. Various strategies, including those discussed above, are employed to address these issues [40–47]. For example, doping cathode materials with elements like niobium (Nb) or aluminum (Al) can enhance the electronic conductivity while maintaining structural integrity [38,40,44]. Additionally, the utilization of advanced characterization methodologies, such as *in situ* electron microscopy, enabled researchers, as demonstrated by Wang et al. [39], to observe and address structural alterations during cycling, offering invaluable insights for material refinement.

4.5. Role of Electrical Properties (Electronic and Ionic Conductivity)

The performance of SSBs is heavily influenced by the electrical properties of their cathode materials. These properties, specifically electronic and ionic conductivity, play a pivotal role in determining the efficiency of charge and ion transport within the battery. Electronic conductivity refers to the ability of a material to conduct electrons. In the context of SSBs, high electronic conductivity ensures that electrons can move freely from the cathode to the anode during the discharge process, and vice versa during charging [48]. Nanostructuring is a technique that has been employed to enhance this property. By reducing the size of the cathode material to the nanoscale, the pathways for electron diffusion become shorter, leading to improved electronic conductivity [49].

On the other hand, ionic conductivity pertains to the movement of ions (in this case, Li ions) within the battery. A high ionic conductivity ensures that Li ions can move swiftly from the anode to the cathode during charging and in the reverse direction during discharging. One way to boost ionic conductivity is by incorporating solid-state electrolytes within the cathode structure. This facilitates faster Li-ion transport, leading to a more efficient battery [49].

A recent advancement in the field of SSBs is the development of the Chevrel Phase Mo_6S_8 nanosheets [50]. These nanosheets are characterized by their high electronic conductivity, rapid ion transport capability, and strong affinity for lithium polysulfides. The unique feature of these nanosheets is their reversible electrochemical Li-ion intercalation. This means that during the battery's operation, the Mo_6S_8 nanosheets can dynamically enhance their ionic conductivity by undergoing a reversible process of lithium-ion intercalation, forming $\text{Li}_x\text{Mo}_6\text{S}_8$. This dynamic enhancement suppresses the undesirable shuttling effect and accelerates the conversion kinetics, leading to superior battery performance in terms of cycling stability, high-rate capability, and low-temperature performance [50].

4.6. Role of Thermal Characteristics (Heat Dissipation and Thermal Stability) in Safety

Thermal characteristics are paramount for the safe operation of SSBs, particularly in high-demand applications such as electric vehicles and large-scale energy storage systems. The inherent risks associated with the thermal behavior of batteries, especially during charging, have led to numerous fire incidents in electric vehicles [51]. Solid-state polymer electrolytes (SPEs) have emerged as a promising solution due to their unique characteristics [52]. Unlike their liquid counterparts, SPEs are not prone to leakage and exhibit low flammability, excellent processability, good flexibility, high safety levels, and superior thermal stability [52]. However, the challenge remains in ensuring that these electrolytes can maintain their stability and performance under various thermal conditions. One of the critical aspects of ensuring thermal safety is understanding and mitigating the heat generated during the battery's operation. For instance, during the charging process, irreversible heat is a significant contributor to temperature rise. Nonetheless, as the battery transitions from a normal charge to an overcharged state, other heat sources, such as reactions due to Mn dissolution and Li deposition, become dominant [51]. Furthermore, the thermal stability of cathode materials is of utmost importance. A study on nickel-based layered cathode materials using *in situ* soft XAS measurements revealed that NiO-type rock salt structures formed at the surface at temperatures above 200 °C, indicating potential thermal

degradation pathways [53]. To ensure the safety of SSBs, it is crucial to have a comprehensive understanding of the thermal behavior of all components. Advanced characterization techniques, such as *in situ* soft XAS and impedance spectroscopy, provide valuable insights into the thermal and electrochemical stability of battery materials [53]. By addressing these thermal challenges, researchers and manufacturers can pave the way for safer and more efficient solid-state batteries.

4.7. Characterization Techniques in the Development of Solid-State Cathodes

The development of solid-state cathodes for advanced battery technologies is a complex and multifaceted process, wherein characterization plays a key role. Characterization techniques are essential in understanding the physicochemical properties of cathode materials, which directly influence the performance, efficiency, and longevity of solid-state batteries. Advanced techniques like X-ray diffraction (XRD), scanning electron microscopy (SEM), and transmission electron microscopy (TEM) provide critical insights into the crystalline structure, morphology, and compositional uniformity of cathode materials [5,13,19]. XRD, for instance, is instrumental in determining phase purity and crystallinity, which are vital for ensuring consistent electrochemical performance. SEM and TEM, on the other hand, offer high-resolution imaging capabilities, enabling researchers to examine the microstructural changes in cathode materials during battery operation, a factor crucial for assessing material stability and cycling behavior [19].

Electrochemical characterization methods such as cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) are equally important in evaluating the performance of solid-state cathodes. These techniques provide valuable information about the redox processes, ion diffusion kinetics, and interfacial properties of the cathode materials [10,38,39]. CV helps in understanding the oxidation and reduction reactions within the cathode, critical for optimizing the energy density and rate capability of the battery. EIS, in particular, is crucial for studying the impedance changes over the battery's lifespan, offering insights into the degradation mechanisms and thereby guiding the development of more durable cathode materials [39].

In addition to structural and electrochemical characterizations, thermal analysis techniques such as differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) are indispensable for evaluating the thermal stability of cathode materials. This aspect is particularly important in the context of solid-state batteries, where thermal management is a key concern [40,45,47]. DSC and TGA can identify the specific temperature ranges where structural changes or decomposition occur, providing vital data for ensuring the safety and reliability of the battery under various operational conditions [40,45]. Comprehensive thermal characterization not only aids in the selection of suitable cathode materials but also informs the design of battery management systems to prevent overheating and thermal runaway.

These characterizations collectively form the backbone of cathode development for solid-state batteries, enabling researchers and engineers to tailor materials for specific applications, enhance performance metrics, and ensure safety and longevity. As the demand for high-performance energy storage solutions grows, the role of thorough and innovative characterization techniques becomes increasingly critical in the quest to unlock the full potential of solid-state battery technology.

The following is a summary and a broader overview of the structural optimization of cathode materials in SSBs: (a) Nanostructuring, which reduces cathode materials to the nanoscale, significantly improves conductivity and the electrochemical reaction surface area. Studies, like those by Sun et al. and Liu et al., have showcased enhanced performance in SSBs using nanostructured cathodes like Li_2WO_4 -coated LiCoO_2 and $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ with a pyrochlore-type $\text{La}_2\text{Sn}_2\text{O}_7$ ceramic layer, respectively. These structures have demonstrated substantial improvements in ionic conductivity and battery performance. (b) Surface coatings: Surface coating of cathode materials with conductive and protective materials addresses issues like structural instability and reactivity with electrolytes. Liang

et al.'s research on a gradient oxy-thiophosphate coating of NMC cathodes showed a reduced capacity fading and an improved cycling stability, enhancing both performance and safety. (c) Composite approaches: Composite cathode materials integrate various components to leverage their complementary properties, enhancing electronic conductivity and ion diffusivity. This approach, seen in the work of Liu et al. and Liang et al., includes strategies like conductive polymer incorporation and gradient coatings for Ni-rich layered oxide cathodes, improving interface and stability. (d) Addressing low conductivity and structural instability: To mitigate fundamental challenges of low electrical conductivity and structural instability, methods like doping with niobium or aluminum and advanced characterization techniques are used. This approach helps in enhancing electronic conductivity and maintaining structural integrity. (e) The role of electrical properties: The electronic and ionic conductivity of cathode materials is crucial for efficient charge and ion transport. Techniques like nanostructuring enhance electronic conductivity by shortening electron diffusion pathways. Incorporating solid-state electrolytes within the cathode structure boosts ionic conductivity, ensuring swift Li-ion movement. (f) The role of thermal characteristics in safety: Thermal characteristics, including heat dissipation and stability, are critical for the safe operation of SSBs. Solid-state polymer electrolytes (SPEs) offer low flammability and superior thermal stability. Understanding and managing the heat generated during operation is essential for safety, especially in high-demand applications. (g) Characterization techniques: The development of solid-state cathodes relies heavily on characterization techniques like XRD, SEM, TEM, CV, EIS, DSC, and TGA. These methods are pivotal in understanding the physicochemical properties, thermal stability, and electrochemical behavior of cathode materials, guiding the optimization for performance and safety.

In terms of future research directions in cathode material optimization for SSBs, we can mention the following. (a) Advanced nanostructuring techniques: Exploring innovative methods in nanostructuring to further enhance the surface area, conductivity, and electrochemical performance of cathode materials. (b) Innovative surface coating materials and methods: Developing new materials and techniques for surface coating to improve cathode stability, reactivity, and overall performance. (c) Synthesis of novel composite cathode materials: Creating composite cathodes with enhanced properties through the integration of conductive polymers, ionic conductors, and other functional materials. (d) Targeted doping strategies: Investigating the effects of various doping elements on the conductivity and stability of cathode materials to enhance their performance. (e) Enhanced characterization and analysis: Utilizing advanced characterization techniques to gain deeper insights into the structural, electrochemical, and thermal properties of cathodes, guiding material refinement. (f) Optimization of thermal management: Focusing on the thermal behavior of cathodes and developing strategies for effective heat dissipation and stability to ensure safety in high-demand applications. (g) Electrolyte–cathode interface engineering: Researching the interfacial interactions between cathodes and solid electrolytes to improve the efficiency and longevity of SSBs. (h) Scalable manufacturing processes: Developing cost-effective and scalable manufacturing techniques for advanced cathode materials to facilitate their widespread application in solid-state batteries.

5. Integration of Advanced Cathode Materials into SSBs

5.1. Scalable Manufacturing Techniques

The industry is continuously seeking innovative and sustainable solutions to enhance battery performance, safety, and manufacturing efficiency. One such innovation is LIOVIX™, a proprietary lithium metal product [54]. This unique printable formulation of lithium metal and other specialty materials can enhance lithium-ion battery performance, reduce manufacturing costs, and pave the way for next-generation battery technology. LIOVIX™ can be printed on a battery's anode during electrode manufacturing in a process called pre-lithiation. This process employs an external source of lithium to offset the first cycle loss of lithium due to SEI (solid electrolyte interphase) formation, allowing for a more

efficient use of lithium from the cathode and increasing the cell energy density. Importantly, LIOVIX™ technology is scalable, using industry-standard coating and printing equipment and can be adapted to various anode or cathode chemistries [54]. Figure 7 shows the cycle performance for commercially available electrode materials with anode loading >4 mAh cm⁻².

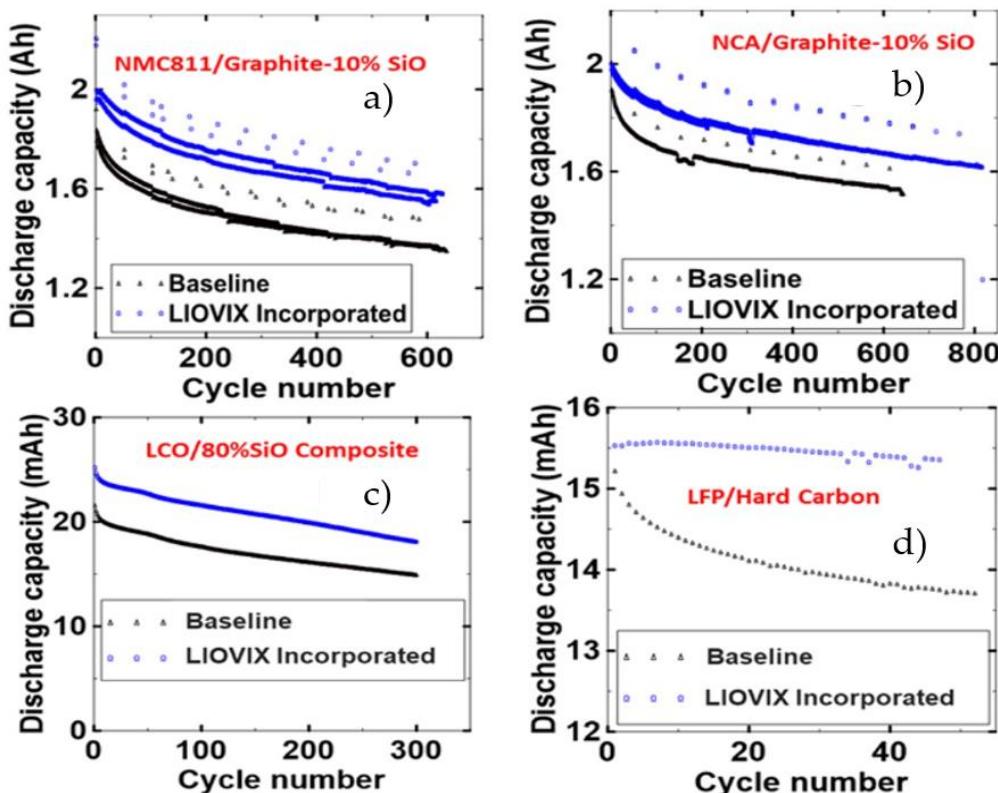


Figure 7. Cycling stability of commercially accessible electrode materials with anode capacities greater than 4 mAh cm⁻². (a) NMC811/Graphite-10@SiO configuration; (b) NCA/Graphite-10%SiO setup; (c) LCO/80%SiO Composite arrangement; (d) LFP/Hard carbon composition (reprinted with permission from ref. [54], Copyright 2022, The Electrochemical Society).

The introduction of LI-OVIX™ into the manufacturing process signifies a major shift in how electrodes are prepared and optimized. Traditionally, the first cycle inefficiency in lithium-ion batteries has been a significant hurdle, leading to a loss in capacity and overall efficiency. The pre-lithiation step facilitated by LI-OVIX™ effectively counteracts this issue by supplementing the anode with additional lithium. This not only improves the first cycle's efficiency, but also enhances the long-term cycle stability of the battery. Studies have shown that pre-lithiated anodes can exhibit a significantly improved cycle life and capacity retention, particularly in high-energy density cells [54]. This aspect of LI-OVIX™ could be particularly advantageous for electric vehicle applications, where both energy density and cycle life are critical parameters.

Furthermore, the compatibility of LI-OVIX™ with various anode and cathode chemistries broadens its applicability across different battery technologies. For instance, its integration into silicon-based anodes, which typically suffer from large volume changes during cycling, could mitigate these effects and enhance the anode's structural integrity [54]. Additionally, when used with high-voltage cathode materials, LI-OVIX™ could potentially stabilize the cathode–electrolyte interface, thereby improving the overall safety and thermal stability of the battery [54]. These advantages position LI-OVIX™ as a versatile tool in the battery manufacturing sector, capable of addressing some of the key challenges faced in the development of advanced lithium-ion batteries.

5.2. Electrode–Electrolyte Interfaces

All-solid-state batteries have the potential for high energy density and enhanced safety due to their nonflammable solid electrolytes. However, a significant challenge is the sluggish ion transmission at the cathode–electrolyte (solid/solid) interface, which results in high resistance at the contact [55]. This limits the practical implementation of these materials in real-world batteries. Several methods have been proposed to enhance the kinetic condition of ion migration between the cathode and the solid electrolyte. One such method is a composite strategy that mixes active materials and solid electrolytes for the cathode, aiming to decrease the ion transmission barrier at the interface [55].

A study presented by Shi and group [55] marks a significant milestone in the evolution of all-solid-state lithium–sulfur (Li–S) batteries, a domain critically important for advancing energy storage technology. The innovative approach of fabricating a solid-state sulfur cathode using a composite of sulfurized polyacrylonitrile (SPAN), lithium bis(fluorosulfonyl)imide (LiFSI), and nanographene wire (NGW) addresses two of the most challenging aspects in solid-state batteries: enhancing interfacial contact and improving both ionic and electronic conduction within the solid cathode structure. This is particularly crucial in solid-state batteries where the absence of a liquid electrolyte can often lead to poor ion transport and interfacial instability. By optimizing the cathode structure, the study contributes significantly to the broader effort of making solid-state batteries a viable alternative to traditional lithium-ion batteries, especially in applications demanding higher energy density and safety. The results showed that at a lower current density of 0.167 mA/cm^2 (0.1C), the solid SPAN/LiFSI/NGW cathode in a bilayer garnet structure achieves an average discharge capacity of 1400 mAh/g over 40 cycles. This high capacity, particularly at a low C-rate, indicates an efficient utilization of active material and minimal capacity fade over the cycles, highlighting the success of the material composition and structural design in maintaining electrochemical stability. Moreover, even at a higher current density of 0.84 mA/cm^2 (0.5C), the cathode maintains a capacity of 437 mAh/g over 200 cycles. This sustained capacity at a higher C-rate is indicative of the cathode's robustness and the effective electron and ion transport within the structure. These performance metrics are particularly noteworthy as they demonstrate the potential of this cathode architecture in real-world applications, where batteries are subjected to varying rates of charge and discharge. Additionally, the operational temperature of 60°C for the battery tests is a critical aspect of the study. The ability of the battery to maintain high performance at this elevated temperature is a promising sign of its suitability for practical applications, where thermal stability is a key concern. Often, solid-state batteries face challenges in maintaining performance at higher temperatures due to the degradation of solid electrolytes and interfaces. The results of this study suggest that the chosen materials and cathode architecture can effectively mitigate these issues, further solidifying the potential of this technology in diverse applications, ranging from electric vehicles to grid storage.

5.3. Current Collectors

In the realm of SSBs, the role of current collectors is paramount for the effective integration of advanced cathode materials [56]. Current collectors serve as the conduit for electron flow, ensuring efficient charge and discharge processes within the battery. However, their significance extends beyond mere electron transport. Recent research [56,57] has highlighted the challenges associated with the interface of current collectors in SSBs, particularly concerning charge transfer kinetics. Deng et al. [56] highlighted the challenge of a limited rate capability as a key barrier to the widespread adoption of inorganic all-solid-state lithium-ion batteries (ASSLIBs). The study pinpointed the importance of charge transfer kinetics at the current collector interfaces for achieving high-rate capacities. To tackle this issue, they proposed the use of a graphene-like carbon (GLC) layer to modify aluminum current collectors. This novel method not only mitigated side reactions at the interface, but also enhanced both the cycling stability and high-rate performance in $\text{LiCoO}_2/\text{Li}_3\text{InCl}_6$ (LCO/LIC) ASSLIBs [56].

Furthermore, the integration of advanced cathode materials in SSBs often necessitates a re-evaluation of the current collector's compatibility and performance [57]. The interplay between the cathode material and the solid electrolyte can introduce challenges at the microstructural, (chemo-)mechanical, and (electro-)chemical levels. Minnmann et al. [57] provided insights into the required properties and potential challenges for inorganic cathode active materials (CAMs) employed in SSBs. They emphasized the importance of tailoring CAMs to address challenges arising from the cathode, particle, and interface levels, particularly concerning the interaction of CAMs with solid electrolytes [57].

5.4. Practical Considerations for Implementing Advanced Cathode Materials

The development and integration of advanced cathode materials are pivotal for the advancement of solid-state battery (SSB) technology, representing a significant leap toward higher energy densities, enhanced safety, and longer cycle life. However, the practical implementation of these materials into SSBs necessitates a comprehensive understanding of both their intrinsic properties and the complex interplay within the battery system, emphasizing the importance of material-specific challenges and system-level considerations. Central to these considerations is the interfacial stability between the cathode active material (CAM) and the solid electrolyte (SE), a critical factor in the performance of SSBs. Achieving a stable interface involves not only ensuring the chemical compatibility between the CAM and SE, but also maintaining their mechanical and thermal stability under operational conditions. Recent studies have focused on the use of interlayer materials that can act as a buffer between the CAM and SE, mitigating interfacial stress and preventing direct contact that could lead to unwanted side reactions [54,55]. Furthermore, advancements in surface modification techniques, such as atomic layer deposition (ALD), have shown promise in creating thin, uniform coatings on CAM particles that enhance ion transport across the interface while protecting the SE from degradation [56].

Despite these advancements, the scalability of protective coatings and interface stabilization techniques presents a significant challenge. Transitioning from laboratory-scale synthesis to industrial-scale production is not straightforward and requires both the upscaling of material fabrication and the retention of material properties at larger scales. This upscaling process must be cost-effective to ensure the economic viability of SSBs. Developing scalable deposition techniques, such as spray pyrolysis or scalable ALD, is essential in this regard [57]. These methods must be optimized to maintain uniformity and quality of the coatings while being efficient and economical for large-scale production.

In addition to material-level considerations, the integration of advanced cathode materials into the overall system design of SSBs is crucial. This includes ensuring compatibility with current collector materials, packaging, and thermal management systems. The design must also account for the mechanical stresses that occur during battery operation, which can affect the integrity of the CAM–SE interface [58,59]. The inclusion of advanced diagnostic tools and sensors in battery systems can provide real-time monitoring of battery health and performance, allowing for the early detection of interface degradation or other issues [58,59].

Addressing these challenges, Ma et al. [13] introduced a novel wet-coating strategy based on preformed nanoparticles to improve interface stability. By employing the non-agglomerated nanoparticles of a coating material, such as ZrO_2 , prepared via solvothermal synthesis, and applying them to a layered Ni-rich oxide CAM after surface functionalization, the authors achieved a uniform surface layer with a unique structure. In pelletized SSBs with argyrodite Li_6PS_5Cl as the SE, the coated CAM exhibited superior lithium storage properties and good rate capability (see Figure 8). The homogeneity of the coating was instrumental in suppressing interfacial side reactions and limiting gas evolution during operation. This strategy proved to be effective not only in SSBs, but also in liquid electrolyte-based Li-ion batteries, demonstrating its potential for broader applications in battery technology [13].

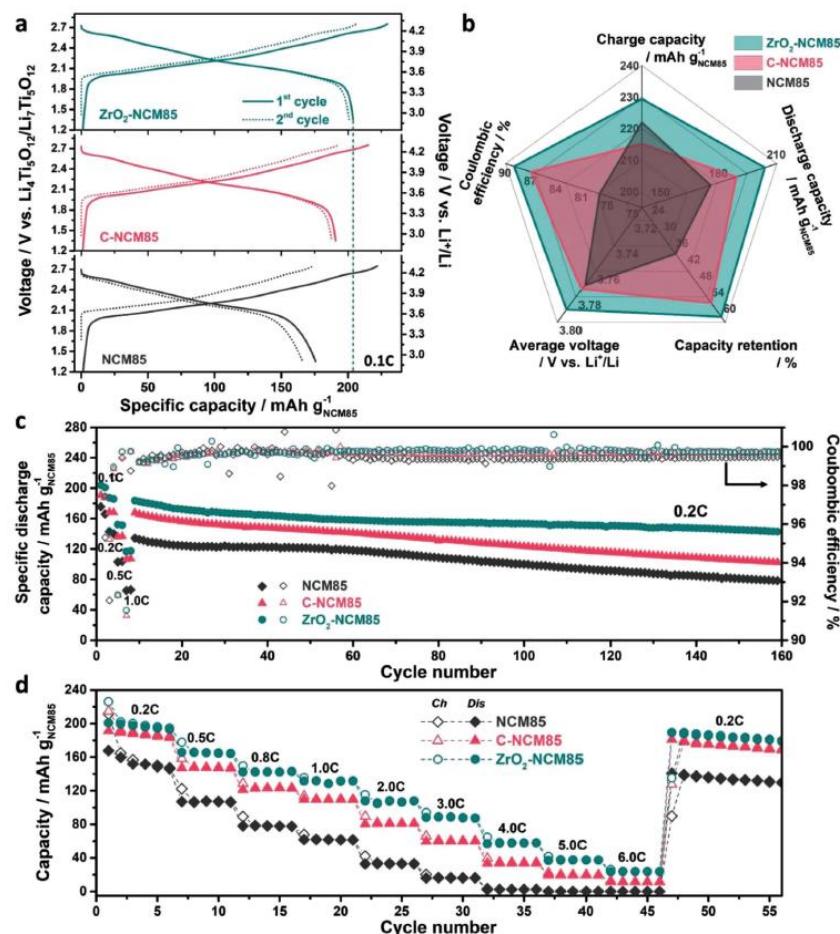


Figure 8. Performance over multiple cycles of bare NCM85, C-NCM85, and ZrO₂-NCM85 in solid-state battery (SSB) cells at 45 °C. (a) Voltage profiles for the first and second cycles at a 0.1C rate. (b) Initial cycle performance metrics and capacity retention calculated by comparing the discharge capacity at 1.0C with the initial discharge capacity at 0.1C. (c) Tests for performance over different rates and long-term cycling. The C-rate ranged from 0.1C to 1.0C for the first 8 cycles, then continued at 0.2C. (d) Specific charge and discharge capacities across various C-rates as a function of cycle number (reprinted with permission from ref. [13], Copyright 2022, John Wiley & Sons).

6. Comparative Evaluation of Cathode Materials

6.1. Key Performance Indicators

The energy density of a battery is a pivotal metric that quantifies the amount of energy a battery can store relative to its volume or weight [56]. This parameter is especially crucial for applications where the constraints of space and weight are paramount, such as in electric vehicles and portable electronics. A battery with a high energy density can store more energy in a smaller space, making it more efficient and desirable for these applications. The high voltage spinel material LiMn_{1.5}Ni_{0.5}O₄ (LMNO) has emerged as a promising candidate to enhance the energy density of lithium batteries [52]. This is attributed to its potential to operate at higher voltages, thereby storing more energy. However, like many advanced materials, LMNO is not without its challenges. Its performance can be compromised by long-term cycling and high-temperature stability. These challenges can lead to a decrease in capacity over time, especially when the battery is subjected to repeated charge and discharge cycles or operated at elevated temperatures [52].

Recent advancements in the field have shown that by partially substituting Mn with Ti in LMNO, the limitations associated with long-term cycling can be effectively addressed [60]. This modification has been shown to enhance the stability of the material,

allowing it to achieve up to 2000 cycles at a high C-rate, which is a measure of the rate at which a battery is discharged relative to its maximum capacity [60].

6.1.1. Voltage Stability

Voltage stability is a critical parameter in battery performance, referring to the battery's ability to maintain a consistent voltage during discharge [28]. A stable voltage is essential as it ensures consistent performance of the battery, preventing potential damage to devices powered by it. A battery that can maintain its voltage during discharge can deliver power more reliably, ensuring that the device it powers operates efficiently and safely.

In the domain of energy storage, supercapacitors have emerged as a promising technology due to their high-power density and long-term durability [61]. A significant advancement in this field is the development of aqueous supercapacitors with extended voltage windows. One such innovation is the development of an aqueous supercapacitor with a high-voltage window of 2.0 V. Liu et al. [61] developed a 2.0 V high-voltage aqueous supercapacitor using core–shell MoO_{3-x} /polypyrrole (MP) nanocomposites as both cathode and anode materials. They contend that the ultra-thin polypyrrole layer on the MoO_3 core not only boosts the conductivity and cycle stability of these nanocomposites, but also serves as a reducing agent, leading to the creation of oxygen vacancies within the MoO_3 core. When utilized as a cathode material, the resultant MP nanocomposite exhibits a potential range extending to 1.0 V. The team concludes that the MP nanocomposite synthesis process is straightforward, and the enhancement in electrode performance is notable, making these nanocomposites promising for high-energy density aqueous supercapacitors (see Figure 9).

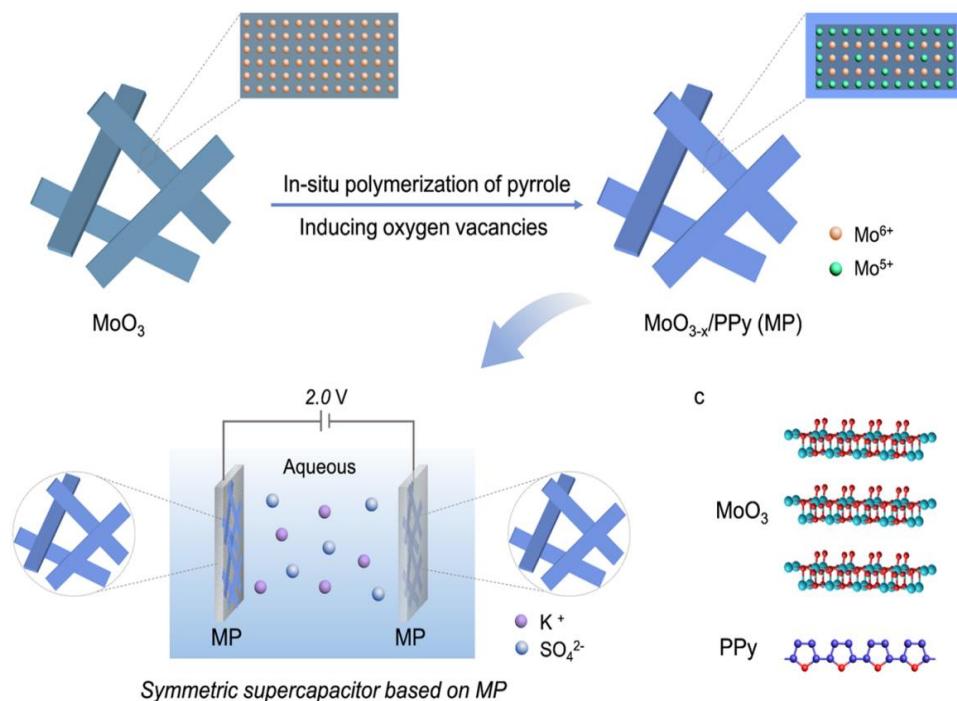


Figure 9. Illustrative summary of the aqueous supercapacitor featuring a 2.0 V high-voltage window using core–shell MoO_{3-x} /polypyrrole (MP) nanocomposites as the material for both the cathode and anode (reprinted with permission from ref. [61], Copyright 2022, American Chemical Society).

6.1.2. Cycle Life

One of the critical performance metrics for batteries is their cycle life. Cycle life is a measure of a battery's longevity and indicates the number of charge and discharge cycles a battery can undergo before its capacity drops to a predetermined percentage of its original value, typically 80% [41]. A battery with a longer cycle life is more desirable as it can be used for more extended periods without significant degradation in performance.

In the quest for high-energy density lithium-ion batteries, high-voltage high-nickel low-cobalt lithium layered oxide cathodes have emerged as potential candidates [62]. These cathodes can store a significant amount of energy, making them suitable for applications that require long-lasting power. However, like many advanced materials, their cycle life can be compromised due to structural deterioration over time. This deterioration can lead to a decrease in capacity and overall battery performance.

Recent research has inquired into strategies to improve the cycle life of these high-nickel cathodes. One such study revealed that doping the cathode material $\text{LiNi}_{0.6}\text{Co}_{0.05}\text{Mn}_{0.35}\text{O}_2$ with a small dose of titanium (Ti) can significantly enhance the cell's performance. This doping strategy not only improved the structural stability of the cathode, but also led to a notable enhancement in its cycle life [62].

7. Identification of Research Gaps and Challenges

7.1. Existing Research Gaps in SSB Cathodes

ASSLBs are increasingly being recognized for their enhanced safety features. To fully harness their capabilities, the integration of high-voltage cathodes is crucial [63–66]. This would elevate the energy density of solid batteries, positioning them as strong contenders against their liquid counterparts. However, the incorporation of high-voltage cathodes is not without its challenges. The stability of the cathode material under high voltages is a significant concern [65]. For instance, in the range of solid-state sodium-ion batteries (SSSBs), the electrochemical stability between sulfide-based solid electrolytes and high-voltage oxide cathodes has been a limiting factor for their long-term performance [65].

The chemical stability at the interface between the cathode and the electrolyte is utmost. Recent studies have highlighted the potential of halide-based solid electrolytes, which demonstrate compatibility with cathodes and exhibit high ionic conductivity [66]. However, even these advanced materials face challenges when it comes to ultra-high voltage operations [66]. The mechanical integrity at the electrode–electrolyte interface is crucial for maintaining a consistent ionic flow and overall battery performance. The generation of gases within the battery can lead to swelling, internal pressure build-up, and potential rupture, compromising the safety and longevity of the battery [66].

To address these challenges, recent research endeavors have been directed toward enhancing solid-state electrolytes [67]. This includes the exploration of polymer solid electrolytes, sulfide solid electrolytes, and oxide solid electrolytes. Furthermore, strategies such as coating protection, synthesis modification, and the structural improvement of cathode materials have been proposed to bolster the electrochemical performance of these batteries [67]. For example, Zhang and colleagues [67] engineered a dual-halogen lithium-ion conductor, namely $\text{Li}_3\text{InCl}_{4.8}\text{F}_{1.2}$. They showcased how fluorine selectively occupies a specific site within the crystal lattice of the solid superionic conductor (Li_3InCl_6), resulting in the formation of a novel dual-halogen solid electrolyte (DHSE). This incorporation of fluorine rendered the $\text{Li}_3\text{InCl}_{4.8}\text{F}_{1.2}$ DHSE denser and capable of sustaining an ionic conductivity above $10^{-4} \text{ S cm}^{-1}$ at room temperature. Additionally, the researchers noted that this DHSE exhibited a practical anodic stability exceeding 6 V (vs. Li/Li^+), facilitating the development of high-voltage all-solid-state lithium-ion batteries (ASSLIBs) with commendable cycling performance. They concluded that their findings offer a novel approach to designing rapid lithium-ion conductors with enhanced oxidative stability, showing considerable promise for application in high-voltage ASSLIBs. Figure 10 shows the electrochemical performance of full cells utilizing $\text{Li}_3\text{InCl}_{4.8}\text{F}_{1.2}$ and Li_3InCl_6 as cathode materials.

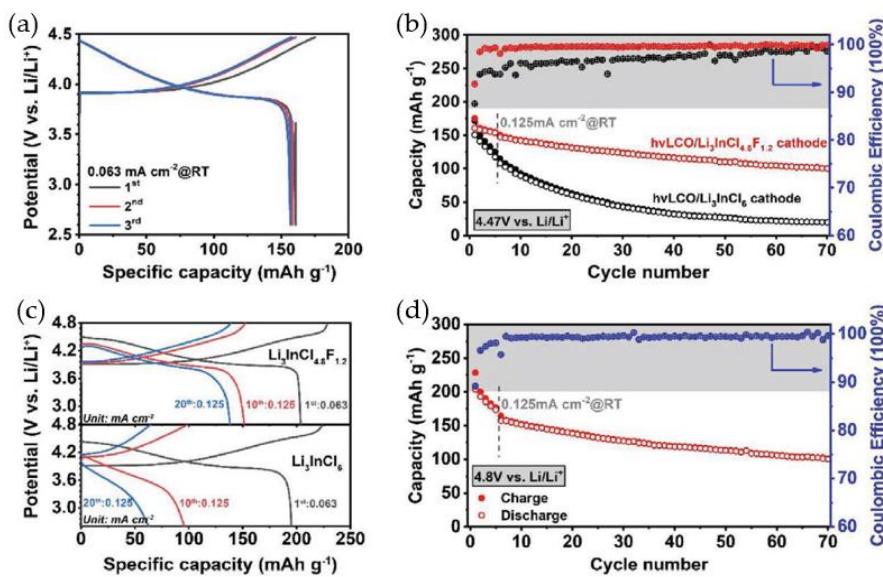


Figure 10. Electrochemical behavior of full cells with Li₃InCl_{4.8}F_{1.2} and Li₃InCl₆ as cathode solid electrolytes (SE). (a) Initial three cycles of discharge and charge for the Li₃InCl_{4.8}F_{1.2} cathode SE cell. (b) Cycle stability of both Li₃InCl_{4.8}F_{1.2} and Li₃InCl₆ cathode SE cells within a voltage span of 2.6–4.47 V (vs. Li/Li⁺) (initial 5 cycles at 0.063 mA cm⁻²). (c) Charge and discharge patterns of Li₃InCl_{4.8}F_{1.2} (upper) and Li₃InCl₆ (lower) cathode SE cells at the 1st, 10th, and 20th cycles in a voltage range of 2.6–4.8 V (vs. Li/Li⁺). (d) Cycle behavior of the Li₃InCl_{4.8}F_{1.2} cathode SE cell when charged to 4.8 V (vs. Li/Li⁺) (first 5 cycles at 0.063 mA cm⁻²) (reprinted with permission from ref. [67], Copyright 2021, John Wiley & Sons).

7.2. Challenges in the Development and Commercialization of SSB Cathodes Metal–Chalcogen Batteries (MCBs)

MCBs are increasingly recognized as potential successors in the realm of energy storage solutions [68,69]. Their cost-effectiveness, impressive theoretical capacity, and environmental compatibility make them stand out. However, the road to their commercialization is riddled with challenges. During the charge and discharge cycles, the electrodes in MCBs undergo significant volume changes. This can lead to mechanical degradation and reduced cycle life of the battery [69]. The dissolution and migration of soluble intermediates, particularly lithium polysulfides, can result in a phenomenon known as the shuttle effect. This not only reduces the battery's efficiency, but also its overall lifespan [69]. The intermediate conversions in MCBs often suffer from sluggish reaction kinetics, which can impede the battery's performance [70]. The growth of uncontrolled dendrites on alkali metal anodes can pose serious safety risks, including short-circuiting and potential battery failure [70]. To address these challenges, the integration of metal organic framework (MOF)-based materials into MCBs has been proposed. MOFs, with their unique properties such as high porosity, low density, expansive surface area, regular pore channels, adjustable pore size, and topological diversity, can effectively mitigate the aforementioned issues. For instance, the anionic Bio-MOF-100 and its derived single-atom zinc catalyst have been shown to simultaneously inhibit lithium dendrite growth and the shuttle effect, leading to enhanced battery performance [71].

8. Future Directions in SSB Cathode Development

The application of coating techniques such as atomic layer deposition (ALD) and physical vapor deposition (PVD) has emerged as a critical strategy in enhancing the performance of ASSBs. ALD, in particular, offers precise control over the thickness and composition of the coatings, enabling uniform coverage of the cathode active material (CAM) at the nanoscale. This uniformity is crucial in reducing the cathode–electrolyte interfacial resistance and in protecting the cathode material from direct contact with the solid electrolyte,

thereby mitigating undesirable side reactions [72]. The use of lithium niobate coatings, for instance, has demonstrated improvements in structural stability and ion transport at the cathode interface, which are pivotal for enhancing the rate capability and cycle life of NCM811-based ASSBs [72].

Laser sintering as a fabrication technique for ceramic-based solid-state battery materials offers several advantages. The method's ability to achieve high heating rates over short interaction durations is particularly beneficial for materials like LLZO and LATP. These elevated heating rates play a crucial role in preserving the crystal structure and enhancing the adhesion of particles, which is vital for maintaining the integrity and ionic conductivity of the solid electrolyte layer [73]. Recent studies have also explored the optimization of laser sintering parameters, including laser power, scanning speed, and layer thickness, to achieve desired microstructural properties in ceramic electrolytes, thus tailoring them for specific battery applications [73,74].

The use of lithium-rich antiperovskites (LiRAPs) as sintering aids represents an innovative approach to lower the sintering temperature of ceramic-based batteries. This reduction in sintering temperature is critical as it minimizes grain growth and preserves the desired microstructure, essential for achieving high ionic conductivity and mechanical strength in solid electrolytes [74]. LiRAPs, with their unique crystal structure and chemical stability, can enhance the sintering process without compromising the electrochemical performance of the resulting solid electrolyte. Further research is needed to fully understand the interactions between LiRAPs and traditional ceramic materials during the sintering process, particularly in terms of phase stability and ionic pathway formation [74].

In addition to novel fabrication techniques, the integration of advanced characterization methods is vital for the progression of ASSB cathode development. Techniques such as *in situ* X-ray diffraction (XRD) and neutron scattering are becoming increasingly important for understanding the structural dynamics of cathode materials under operational conditions [75]. These methods allow for real-time observation of phase transformations, lattice expansions, and potential microstructural changes during battery cycling. Additionally, advanced electron microscopy, including cryogenic transmission electron microscopy (cryo-TEM), offers unparalleled insights into the nanoscale architecture of coated cathode materials, revealing interface phenomena that are crucial for optimizing material design and performance [75]. The combination of these sophisticated characterization tools with computational modeling provides a holistic approach to material analysis, enabling the prediction and tailoring of material properties to meet specific performance criteria, such as enhanced ionic conductivity and mechanical stability [75].

9. Evolution of All-Solid-State Battery Technology: Key Milestones and Developments from 1990 to 2022

The evolution of all-solid-state batteries from the 1990s to this day marks a significant paradigm shift in energy storage technology, highlighting the transition from traditional lithium-ion systems to safer, more efficient alternatives. Key milestones, such as the development of high ionic conductivity solid electrolytes and the integration of lithium metal anodes, have been pivotal in enhancing energy densities and addressing safety concerns inherent in conventional batteries. These advancements not only underscore the rapid progression in battery technology, but also set the stage for the next generation of high-performance reliable energy storage solutions, crucial for a wide range of applications. Table 2 presents a scheme of the evolution of solid-state batteries from 1990 to 2022.

Table 2. Scheme of the evolution of solid-state batteries from 1990 to 2022.

Year/Period	Development Milestone	Key Examples/Notes	Reference
Early 1990s	Initial exploration of solid electrolytes, focusing on stability and ionic conductivity.	Introduction of lithium phosphorus oxynitride (LiPON) and thio-LISICON as potential solid electrolytes.	[76]
Mid-1990s	Development of polymer-based solid electrolytes for enhanced flexibility and processability.	Advancements in poly(ethylene oxide) (PEO)-based electrolytes, demonstrating improved electrochemical stability.	[77]
Late 1990s	Emergence of oxide-based solid electrolytes, offering improved ionic conductivity and stability.	Lithium lanthanum titanate (LLTO) and other oxide ceramics explored for higher conductivity and stability.	[78]
Early 2000s	Introduction of sulfide-based solid electrolytes, significantly enhancing ionic conductivity.	LGPS ($\text{Li}_{10}\text{GeP}_2\text{S}_{12}$) showing high ionic conductivities comparable to liquid electrolytes.	[79]
2007	Breakthrough in garnet-type solid electrolytes with the development of $\text{Li/La}_3\text{Zr}_2\text{O}_{12}$ (LLZO).	LLZO noted for its high ionic conductivity, stability against lithium metal, and low grain boundary resistance.	[80]
Late 2000s	Progress in thin-film solid-state batteries, focusing on integration into electronic devices and miniaturization.	Development of micro-batteries for electronic devices, leveraging thin-film technology for compact design.	[81]
Early 2010s	Advances in interface engineering between solid electrolytes and electrodes to enhance performance and durability.	Exploration of various coating and interface modification techniques to reduce interfacial resistance and improve cycling stability.	[82]
2012	Development and adoption of composite solid electrolytes combining polymers and ceramics for improved properties.	Composite electrolytes exhibiting enhanced mechanical properties and ionic conductivity, suitable for practical applications.	[83]
Mid-2010s	Scale-up and design improvements for larger applications, particularly in the electric vehicle sector.	Transition from laboratory-scale research to prototypes demonstrating feasibility for electric vehicle applications.	[84]
2017	Surge in commercial interest and investments for the development of solid-state batteries in electric vehicles.	Major automotive and battery manufacturers announcing projects and collaborations for ASSB development.	[85]
2018	Addressing lithium dendrite formation in solid-state lithium metal batteries.	Innovations in electrolyte and anode design to suppress dendrite growth, enhancing safety and cycle life.	[86]
2020	Advancements in scalable manufacturing techniques for solid-state batteries, focusing on cost reduction and mass production.	Development of novel manufacturing processes, including roll-to-roll and sputtering techniques for large-scale production.	[87]
2021	Enhanced understanding and mitigation of interfacial challenges to improve cycle life and performance.	Research on interfacial phenomena, leading to strategies for stabilizing the electrode/electrolyte interface.	[88]
2022	Introduction of novel materials and hybrid systems, pushing the boundaries of energy density and safety.	Exploration of new solid electrolyte materials and innovative hybrid solid–liquid electrolyte systems for higher performance batteries.	[89]

10. Conclusions

In conclusion, our review underscores the transformative potential and ongoing challenges in the development of SSB cathodes. The future direction of SSB cathode design is geared toward addressing these challenges, paving the way for more efficient, safe, and sustainable energy storage solutions. A summary of the most import points of the review is presented below:

Global Energy Shift and SSBs' Role:

- The global transition from fossil fuels to cleaner energy alternatives has heightened the need for high-performance energy storage systems.
- SSBs emerge as a promising successor to conventional lithium-ion batteries, offering enhanced energy density, superior safety, and extended service life.

Foundational and Novel Cathode Materials:

- Traditional cathode materials like LiCoO_2 , LiMn_2O_4 , and LiFePO_4 have been instrumental in advancing SSB technology.
- Novel cathode materials, including sulfides, oxides, and air-based cathodes, present unique benefits and challenges, particularly in their interaction with solid electrolytes.

Strategies for Cathode Optimization:

- Key strategies for improving cathode performance include nanostructuring, surface modifications, and composite formulations.
- These approaches aim to address issues such as limited conductivity and structural vulnerabilities.

Critical Factors for Cathode Performance:

- Electronic and ionic conductivity, along with thermal attributes like heat dissipation and robustness, are crucial for battery safety and efficiency.

Manufacturing and Practical Application:

- Scalable fabrication methods are needed for the successful integration of advanced cathode materials into SSBs.
- Focus on electrode–electrolyte interfaces and current collectors is essential for practical deployment.

Comprehensive Assessment of Cathode Materials:

- A holistic evaluation based on performance metrics such as energy density, voltage stability, and cycle longevity is vital for understanding potential and areas for refinement.

Future Insights and Research Directions:

- Despite significant progress, there are persistent research gaps and challenges in the evolution of SSB cathodes.
- Future research is optimistic, with potential breakthroughs in cathode materials and methodologies.
- The advancement of SSB technology will play a pivotal role in achieving sustainable energy goals.

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