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Innovative Research on Solid-State Electrolyte Materials for Solid-State Batteries

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Abstract. Solid-state batteries, utilizing solid electrolytes, are emerging as a promising next-generation energy storage technology due to their superior safety, energy density, and lifespan. This study investigates key performance bottlenecks in solid-state electrolytes, with a focus on enhancing ionic conductivity, interfacial stability, and maneuverability for large-scale industrial applications. We conduct a comparative analysis of ion conduction mechanisms, structural characteristics, and interfacial behaviors across different types of solid-state electrolytes—oxide, sulfide, polymer, and hairslide systems. Current solid electrolyte systems exhibit diverse properties and limitations. Polymer electrolytes show ionic conductivity in the range of ~10^{-7}–10^{-5} S·cm^{-1}, while sulfide electrolytes range from ~10^{-3}–10^{-2} S·cm^{-1}. However, interface resistances often exceed 100 Ω·cm^2 without surface modifications. Significant progress has been made in hybrid and interface engineering, such as LLZO polymer composites and sulfide@oxide coatings, which reduce contact resistance by 40–70%. An ideal electrolyte system capable of balancing high ionic conductivity, chemical stability, and maneuverability remains elusive. To facilitate industrialization, critical challenges such as processing temperature limits, air sensitivity, and cost-effective material handling must be addressed.

Keywords: Solid-state electrolytes, ionic conductivity, inter facial stability, hybrid systems, solid-state batteries

1. Introduction

The global transition toward sustainable and low-carbon energy systems is driving unprecedented demand for high-performance energy storage technologies. With the rapid expansion of renewable energy deployment and the electrification of transportation, batteries offering higher energy density, improved safety, and longer cycle life have become a central focus of both research and industrial investment. Among the emerging candidates, solid-state batteries (SSBs) are regarded as one of the most promising solutions because of their potential to combine high specific energy with enhanced operational safety.

Conventional lithium-ion batteries (LIBs) employ flammable liquid electrolytes that are vulnerable to leakage, combustion, and dendrite penetration during high-rate charging, thereby posing significant safety hazards [1,2]. In contrast, replacing liquid electrolytes with solid-state electrolytes (SSEs) not only mitigates these risks but also enables the use of lithium-metal anodes, which can further enhance energy density [3].

Several countries and regions—including the United States, Japan, South Korea, and Europe—have launched strategic programs to accelerate the development and industrialization of SSB technologies [2]. China has also made considerable progress, supported by strong academic output and patent activity. Nevertheless, critical challenges remain in raw material supply, interfacial engineering, scalable manufacturing, and ecosystem development [2]. Addressing these issues will be essential for enabling large-scale commercialization and achieving global competitiveness in the SSB sector.

1.1. Research problem

Despite significant progress in the development of various SSE systems—including oxides, sulfides, polymers, and Halogen—no single material currently meets the combined requirements for high ionic conductivity, wide electrochemical stability, robust interfacial compatibility, and cost-effective scalability [3,4].The main technical bottlenecks can be summarized as follows: Limited Li^+ ionic conductivity at ambient temperatures. Electrochemical stability mismatch between electrolytes and high-voltage cathodes or lithium-metal anodes [4]. Interfacial resistance growth due to poor solid–solid contact and side reactions [3]. Environmental sensitivity of certain SSEs, such as sulfides, to moisture and oxygen [2,3]. Processing and cost barriers arising from high sintering temperatures and limited raw material availability [2]. These interrelated issues mean that improving one parameter often comes at the expense of another, making it necessary to adopt integrated design strategies.

1.2. Objectives of the study

This research aims to explore and analyze innovative developments in solid-state electrolyte materials to overcome performance bottlenecks. The specific objectives are:

Investigate how structural and compositional factors influence ion transport in SSEs.

Compare the electrochemical performance of major SSE categories, highlighting trade-offs and optimization paths [3].

Explore engineering strategies for stabilizing electrode–electrolyte interfaces, particularly for high-voltage and lithium-metal configurations [4].

1.3. Significance of the study

A systematic evaluation of SSE materials from both a performance and manufacturing perspective will provide valuable guidance for the development of next-generation SSBs. By integrating material optimization with interface engineering, it is possible to design systems that meet the dual requirements of high conductivity and long-term cycling stability, thereby enabling safer, more efficient, and commercially viable energy storage solutions [2-4].

1.4. Hypothesis

It is hypothesized that composite SSE structures—combining a highly conductive phase with a chemically stable phase—can simultaneously achieve fast Li^+ transport and robust interfacial stability. By tailoring microstructural features such as grain boundary connectivity and phase distribution, and by optimizing interface chemistry, it should be possible to achieve room-temperature ionic conductivities $\geq 10^{-3}$ S·cm^{-1} and interfacial resistances ≤ 50 Ω·cm^2 against high-voltage cathodes, without imposing excessive environmental control requirements [3,4].

1.5. Overview of the paper structure

This paper is organized as follows. Section 2 outlines the ion-conduction mechanisms underlying solid-state electrolytes (SSEs) and clarifies the evaluation metrics used in this review. Section 3 surveys oxide-, sulfide-, polymer-, and halogen-based electrolytes, providing a comparative analysis of their electrochemical, mechanical, and processing characteristics. Section 4 examines interfacial stability at both cathode/SSE and Li-metal/SSE contacts and summarizes state-of-the-art engineering strategies. Section 5 synthesizes recent advances, identifies unresolved bottlenecks, and maps research gaps to promising

2. Literature review

2.1. Overview of the existing research

Solid-state electrolytes (SSEs) are lithium-ion-conducting materials in solid form, designed to replace liquid electrolytes in conventional lithium-ion batteries and potentially enhance performance and safety. Current SSE systems are typically categorized into four groups: oxide-based, sulfide-based, polymer-based, and Halogen-based electrolytes [2-4].

2.1.1. Oxide-based electrolytes

Representative examples include lithium lanthanum zirconium oxide (LLZO) and lithium aluminum titanium phosphate (LATP). These materials exhibit excellent chemical stability and tolerance to harsh environments. However, they generally suffer from lower ionic conductivity compared to sulfide-based SSEs and have poor interfacial contact with electrodes, which increases interfacial resistance and decreases battery efficiency [2].

2.1.2. Sulfide-based electrolytes

Representative materials of sulfide-based electrolytes include Li10GeP$_2$S$_{12}$ (LGPS) and Li6PS5Cl, etc. Due to their unique crystal structure design, they exhibit extremely excellent lithium-ion conductivity in current solid-state electrolyte systems. Compared with other types of electrolytes, sulfide-based electrolytes perform most prominently in terms of ion conduction. Their electrical conductivity at room temperature can reach 10^{-2} to 10^{-3} S/cm, approaching or even partially surpassing the level of traditional liquid electrolytes. However, this type of electrolyte is extremely sensitive to environmental conditions and readily reacts with moisture in the air to form toxic hydrogen sulfide gas (H_2S), posing a severe challenge to operational safety [3,5].

2.1.3. Polymer-based electrolytes

Common matrix materials include polyethylene oxide (PEO) and polyvinylidene fluoride (PVDF), etc. The lithium ion conduction mechanism is closely related to the thermal motion of the polymer chain segments. Lithium ions usually form transient complex structures by coordinating with polar groups on the polymer molecular chain (such as ether oxygen or fluorine atoms), and the ion migration process is achieved along with the dynamic movement of the polymer chain segments [2].

Polymer-based electrolytes have excellent flexibility and film-forming properties, with mechanical properties close to those of plastic materials. During processing, they can be prepared in various flexible ways such as hot pressing and coating. They are particularly suitable for preparing ultra-thin and bable flexible solid-state batteries and are widely used in wearable electronics, flexible electronic devices and other application scenarios with high requirements for morphological adaptability. However, the ionic conductivity of this type of material at room temperature is generally low. Usually, it needs to be heated above 60°C to activate the effective movement of polymer chain segments, thereby achieving acceptable electrical conductivity. This characteristic to some extent limits its wide application under normal temperature conditions [2].

2.1.4. Halogen-based electrolytes

Halogen-based electrolytes are a new type of electrolyte material that has emerged in recent years, featuring unique crystal structures and excellent interfacial stability. Representative materials include Li_3YCl6, etc. It shows stronger tolerance to oxygen and moisture in the air, can be synthesized and processed under relatively loose environmental conditions, has better process compatibility, and demonstrates good chemical stability when paired with high-voltage cathode materials such as $LiCoO_2$, NCM, etc.

Although the research on halogen-based electrolytes started relatively late, they have demonstrated excellent comprehensive performance and application prospects in the laboratory stage, attracting extensive attention from both the academic and industrial communities. Current research mainly focuses on enhancing its electrical conductivity, optimizing interface compatibility with electrode materials, and reducing preparation costs. Due to its advantages in safety, environmental stability and compatibility with high-voltage systems, Halogen electrolytes are considered to have significant development potential in application scenarios with high requirements for stability and safety, such as electric vehicles and energy storage systems [4].

2.2. Identification of gaps in the literature

Despite notable advances, each SSE category presents distinct limitations. Oxide-based electrolytes face low conductivity and interfacial incompatibility; sulfide-based materials require strict environmental control; polymer-based electrolytes lack sufficient room-temperature conductivity; and Halogen-based electrolytes are still in early research stages with moderate conductivity. This underscores the necessity of developing materials that simultaneously achieve high conductivity, interfacial stability, manufacturability, and environmental tolerance [2–4].

Hybrid SSEs—combining features such as the high conductivity of sulfides, the stability of oxides, and the flexibility of polymers—could offer a path toward overcoming these limitations.

2.3. Relevance to the current study

This work systematically analyzes ion conduction mechanisms, structural properties, and interfacial behaviors of representative SSE systems. The aim is to identify material design strategies capable of resolving the above challenges and to provide theoretical guidance for advancing solid-state battery performance.

2.4. Theoretical framework

Ion conduction in SSEs can be described by three principal mechanisms, though in practice these often coexist and interact:

2.4.1. Vacancy mechanism

Lithium ions migrate through vacant lattice sites; this mechanism is common in oxides but can also occur in doped Halogen.

2.4.2. Interstitial mechanism

Lithium ions diffuse via interstitial sites; often observed in sulfides, but also relevant in disordered Halogen.

2.4.3. Cooperative mechanism

The cooperative mechanism involves the collective "string-like" motion of multiple ions, which is facilitated by flexible frameworks, sublattice disorder, and rotational entropy.. This mechanism underpins superionic conduction in argyrodites and some halogens, and can be tuned via aliovalent doping and lattice softness [1,4].

These mechanisms form the theoretical foundation for evaluating ionic conductivity and guiding the structural optimization of high-performance SSEs.

3. Results

3.1. Presentation of key findings

Solid-state electrolyte (SSE) systems differ significantly in their electrochemical, mechanical, and processing characteristics, and no single material family currently optimizes all critical performance parameters simultaneously. Here, representative data from the literature [2-4] are used to support a comparative analysis.

3.1.1. Ionic conductivity (σLi^{+}@25 C)

Ionic conductivity is a crucial performance metric for SSEs, with values differing across material types. Sulfides, such as LGPS and argyrodites, show high ionic conductivity (10^{-3} to 10^{-2} S·cm^{-1}), which is comparable to liquid electrolytes. Halogen, including Li_3YCl_6 and Li_3InCl_6, exhibit moderate conductivity (10^{-4} to 10^{-3} S·cm^{-1}), while oxides like LLZO demonstrate lower conductivity (10^{-4} to 10^{-3} S·cm^{-1}), which decreases further at grain boundaries. Polymers, such as PEO–LiTFSI, have the lowest conductivity (10^{-7} to 10^{-5} S·cm^{-1}) and require elevated temperatures to achieve higher conductivity.

3.1.2. Chemical and environmental stability

Chemical and environmental stability is another critical factor. Oxides are highly stable in air and moisture, remaining unchanged for months under ambient conditions. Halogen offer better air stability than sulfides but are still prone to hydrolysis over time; moisture-resistant processing is required. Sulfides, on the other hand, rapidly hydrolyze in ambient conditions, releasing H_2S. Polymers are moderately stable, though oxidative degradation may occur over time.

3.1.3. Electrochemical stability (separate eox vs ered)

In terms of electrochemical stability, oxides and halogens exhibit high oxidative stability (Eox vs $Li^+/Li > 4.3–4.5$ V), whereas sulfides and polymers show lower stability (2.5–3.8 V). Most SSEs are thermodynamically unstable against Li^0, with sulfides and Halogen requiring inter phase engineering to suppress reduction. Some polymer-salt systems, however, show improved compatibility with lithium.

3.1.4. Mechanical properties

The mechanical properties of these materials also vary. Polymers are highly flexible, allowing for conformal contact with electrodes. Oxides are brittle and have a high modulus, while sulfides and halogens possess an intermediate modulus and can deform under pressure.

3.1.5. Inter facial compatibility

Interfacial compatibility is another key factor in SSE performance. Polymers, with their low modulus, enable intimate contact with lithium metal, reducing interfacial resistance. Oxides require high-pressure densification to achieve good contact, and sulfides are chemically reactive with high-voltage cathodes, often necessitating coatings. Halogen show better stability at the cathode interface but still face challenges with the Li-metal interface.

3.1.6. Processing complexity

The processing complexity for these materials varies significantly. Sulfides require high handling complexity, including dry-room environments and inert atmosphere milling. Oxides need sintering at temperatures above 1000 C in controlled atmospheres. Halogen have moderate processing complexity, with low-temperature synthesis being possible but dry-room conditions recommended. Polymers are easier to process, requiring solution casting or melt pressing at temperatures below 200 C, though the costs of solvents and salts can be a limiting factor.

3.1.7. Cost considerations

Cost considerations are crucial in the adoption of SSE materials. Raw materials for sulfides (Li_2S) and rare-earth Halogen (Re Cl_3) are expensive, while LLZO precursors are moderately priced. Polymer monomers are inexpensive, but Li salts can be costly. Processing costs for oxides and sulfides are higher due to the need for high-temperature sintering or dry-room conditions. Polymer processing, in contrast, involves lower energy costs. Indicative electrolyte sheet costs range from ~$20 to $80 per square meter, depending on the material and processing methods [5,6].

3.2. Use of tables, figures, and charts

Based on the comparative data from the literature [7,8], the performance of different SSE systems is summarized in Table 1.:

Table 1. A comprehensive comparison of different material systems

Performance indicators	Oxide group	Sulfide group	Polymer group	Halogen group
σLi^+ (25 C, $S \cdot cm^{-1}$)	$1\times10^{-4} - 1\times10^{-3}$	$1\times10^{-3} - 1\times10^{-2}$	$1\times10^{-7} - 1\times10^{-5}$	$1\times10^{-4} - 1\times10^{-3}$
Eox (V vs Li^+/Li)	4.0–4.3	2.5–2.8	3.8–4.0	4.3–4.5
Ered (V vs Li^+/Li)	0.0 – 0.5	0.0 – 1.7	0.0 – 1.0	0.0 – 1.8
interfacial resistance to NMC @ 60 C ($\Omega \cdot cm^2$)	100–500	20–100	10–50	30–120
Ambient handling tolerance	Days–weeks	Minutes	Hours–days	Days
Typical process T (°C)	1000–1250	200–300	60–100	500–600
Powder sensitivity	Low	High (moisture)	Low	Medium (moisture)
Indicative cost ($/m² electrolyte (sheet)	20–40	40–80	5–15	25–50

3.3. Statistical analysis

Quantitative comparison confirms that sulfides lead in conductivity but require strict environmental control and incur high handling costs. Halogen combine moderate conductivity with good high-voltage stability, yet they are limited by raw material expenses and moisture sensitivity. Oxides excel in long-term stability but demand high-temperature processing, while polymers are the most cost-effective to process but suffer from low σLi^+ at room temperature; oxides excel in stability but need high-T processing; polymers are cheapest to process but suffer from low σLi^+ at room temperature. These findings reinforce the importance of hybrid designs that combine complementary strengths [6-8].

4. Discussion

4.1. Summary of key findings

This study comprehensively compared the shortcomings and advantages of solid-state electrolytes (SSEs) systems in terms of structure, performance and application conditions. Oxide-based solid electrolytes are renowned for their excellent chemical stability, but the problem of relatively large interface impedance still needs to be effectively addressed. Sulfide-based solid electrolytes have a relatively high ionic conductivity, significantly enhancing battery performance. However, the strict requirements for the production environment pose considerable challenges to their industrialization. Polymer-based solid electrolytes have good mechanical flexibility and processing advantages, but the limitations of their electrical conductivity at room temperature need to be overcome as soon as possible. Halogen solid electrolytes show relatively balanced performance and have broad prospects, but their long-term stability and large-scale preparation processes still need further verification. These findings support the hypothesis (H1) that composite SSEs structures with mixed materials may offer better overall performance.

4.2. Comparison with previous studies

Previous studies have shown that no electrolyte material can meet all the performance requirements of solid-state batteries, and our research also confirms this view. In traditional systems, ionic conductivity, chemical stability and processing performance are often difficult to achieve simultaneously. Therefore, in recent years, the academic community has increasingly focused on mixed electrolyte systems, which undoubtedly represents a reasonable advancement towards integrating the advantages of different materials to enhance overall performance. For instance, by combining oxides with sulfides, a mixed solid electrolyte with high ionic conductivity and good stability can be obtained. This further confirms that the hybrid electrolyte system is expected to become an important way to achieve the commercialization goal of all-solid-state batteries.

4.3. Implications of the findings

The results show that the hybrid electrolyte system is a feasible way to improve the commercial feasibility of solid-state batteries. By integrating the excellent properties of different electrolyte materials, it is expected to overcome the limitations of a single electrolyte material. However, this requires further progress in interface engineering and material synthesis. These findings suggest that future development should prioritize optimizing interface stability to ensure the stability and safety of long-term cycling.

4.4. Limitations of the study

This study is based on a review of previous solid electrolyte literature and does not include experimental validation data. While some suggestions are provided for optimizing the overall performance of solid-state batteries and promoting their commercialization, empirical research is needed to confirm them. Furthermore, this study does not encompass all possible real-world environmental conditions. Further experimental research is needed to validate these theoretical results under real-world conditions.

4.5. Suggestions for future research

4.5.1. Improving the room-temperature ionic conductivity of SSEs

Future research should focus on developing novel electrolyte materials through strategies such as multi-element doping, microstructure manipulation, and composite material design. These efforts aim to further surpass the limits of room-temperature ionic conductivity (e.g., reaching 10^{-2} S/cm) while maintaining high stability. Furthermore, these efforts should broaden the electrochemical window to accommodate high-energy-density cathode and anode materials.

4.5.2. Optimizing electrolyte interface stability

Future research is recommended to focus on interface engineering strategies, such as developing functional interface buffer layers, designing electrolytes with gradient compositions, or surface modification of electrode materials to improve physical contact, inhibit dendrite growth, and create chemically and electrochemically stable interfaces, thereby significantly improving battery cycle life and safety.

4.5.3. Explore innovative preparation processes

Future research should focus on innovative fabrication processes, striving to develop technological pathways that are lower-cost, environmentally friendly, and easily scalable, paving the way for solid-state batteries to move from the laboratory to the market.

5. Conclusion

This research demonstrates that solid-state batteries, as the core of next-generation energy storage technology, require innovative electrolyte materials with significant and far-reaching implications for advancing the new energy sector. The development of composite electrolyte materials can help maximize the advantages of different solid-state electrolyte materials by leveraging their strengths and minimizing their weaknesses. Furthermore, various optimization strategies have already provided promising approaches for improving interfacial stability, but further research and engineering effort are needed to refine these strategies.

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