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RESEARCH PROGRESS ON SOLID-STATE ELECTROLYTES AND THEIR MANUFACTURING PROCESSES

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Abstract: With the increasing demand for high energy density and enhanced safety in energy storage systems, conventional lithium-ion batteries employing liquid electrolytes have gradually exhibited intrinsic limitations in both safety and performance improvement. All-solid-state batteries, which replace flammable liquid electrolytes with solid electrolytes, are widely regarded as one of the most promising candidates for next-generation high-safety energy storage technologies. However, solid electrolytes still face considerable challenges in terms of material properties, interfacial stability, and large-scale manufacturing, which significantly hinder their practical application. This thesis presents a systematic review of solid electrolytes and their production processes. Firstly, the current research status of major solid electrolyte material systems, including oxide-based, sulfide-based, polymer-based, and composite solid electrolytes, is comprehensively summarized, with particular emphasis on their ionic conduction mechanisms, chemical stability, and engineering applicability. Subsequently, the progress in key fabrication techniques of solid electrolytes is reviewed, focusing on the influence of processing conditions on microstructure and electrochemical performance. Furthermore, the critical interfacial issues in all-solid-state batteries are discussed, and recent advances in interfacial engineering strategies are analyzed. Finally, combined with the current status of industrialization, the technical routes and future development trends of all-solid-state batteries are evaluated. Through an integrated analysis of material systems, manufacturing processes, interfacial engineering, and industrialization pathways, this work aims to provide a systematic reference for research on solid electrolytes and all-solid-state batteries, as well as insights into their further engineering implementation.

Keywords: Solid electrolyte; All-solid-state batteries; Material systems; Manufacturing processes; Interfacial engineering; Industrialization

1 INTRODUCTION

Against the background of the global energy structure transition and the “dual-carbon” strategy, electrochemical energy storage technologies have become increasingly important in new energy vehicles, large-scale energy storage systems, and portable electronic devices. As the most mature energy storage technology at present, lithium-ion batteries have achieved remarkable progress in terms of energy density, cycle life, and manufacturing cost. However, due to the inherent safety risks associated with flammable liquid electrolytes, further improvements toward higher energy density and enhanced safety are constrained by both physical and chemical limitations [1–3].

By replacing conventional organic liquid electrolytes with solid-state electrolytes (Solid-State Electrolyte, SSE), solid-state batteries fundamentally eliminate the risks of electrolyte leakage and thermal runaway, and are therefore regarded as one of the key technological pathways toward next-generation batteries with high safety and high energy density [4–6]. Theoretical studies indicate that solid-state batteries can be directly coupled with lithium metal anodes, significantly increasing both gravimetric and volumetric energy density, with theoretical values exceeding 400 $\mathrm{Wh{\cdot}kg^{-1}}$ [1,7]. In this context, solid-state electrolytes, as the core functional materials of solid-state batteries, play critical roles in lithium-ion conduction, electronic insulation, and electrode separation. Their material system selection, ion transport mechanisms, interfacial stability, and scalable manufacturing processes directly determine the overall performance and industrial feasibility of solid-state batteries [8–10]. This paper aims to systematically review the current research status of solid-state electrolytes and their production processes, and to discuss interfacial engineering strategies, industrialization progress, and future development trends, providing references for further optimization of solid-state battery performance and large-scale application.

2 CLASSIFICATION AND RESEARCH STATUS OF SOLID-STATE ELECTROLYTE MATERIAL SYSTEMS

As the core functional materials of all-solid-state batteries, the selection of solid-state electrolyte material systems directly determines ionic transport capability, electrochemical stability, safety, and engineering feasibility. At present, solid-state electrolytes can generally be classified into three main categories: inorganic solid-state electrolytes, polymer solid-state electrolytes, and composite solid-state electrolytes [2,6,11]. Different material systems exhibit distinct characteristics in terms of ion conduction mechanisms, interfacial behavior, and processing technologies, giving rise to multiple parallel technological development routes.

Inorganic solid-state electrolytes, owing to their relatively high lithium-ion conductivity and excellent thermal stability, are considered one of the most promising candidates for achieving high-energy-density all-solid-state batteries [6,8].

According to their chemical composition and crystal structure, they can be mainly divided into oxide-based, sulfide-based, and halide-based solid electrolytes.

Oxide-based solid-state electrolytes have attracted extensive attention due to their outstanding chemical stability, wide electrochemical windows, and good air stability. Among them, garnet-type $Li_7La_3Zr_2O_{12}$ (LLZO) and NASICON-structured electrolytes are the most extensively studied representative systems [11-12]. Studies have shown that elemental doping (such as Al, Ta, Nb, etc.) can effectively stabilize the cubic phase of LLZO and significantly enhance its room-temperature lithium-ion conductivity, reaching values on the order of 10^{-3} $S \cdot cm^{-1}$ [13]. However, oxide solid electrolytes generally suffer from large grain boundary resistance, poor interfacial contact, and high sintering temperatures, which result in relatively high interfacial resistance in practical batteries [14-15]. In addition, interfacial reactions and lithium dendrite penetration may still occur at the interface between oxide electrolytes and lithium metal anodes, necessitating mitigation through interfacial engineering and structural design [16].

Sulfide-based solid-state electrolytes are regarded as one of the most promising systems for early commercialization due to their extremely high ionic conductivity and favorable interfacial wettability [6,17]. Typical representatives include $Li_{10}GeP_2S_{12}$ (LGPS) and its derivatives, as well as argyrodite-type materials such as Li_6PS_5X (X = Cl, Br, I). Research has demonstrated that sulfide solid electrolytes can achieve room-temperature ionic conductivities up to 10^{-2} $S \cdot cm^{-1}$, approaching or even surpassing those of conventional liquid electrolytes [17]. Moreover, their relatively low Young's modulus facilitates intimate electrode/electrolyte contact, thereby significantly reducing interfacial resistance [6,10]. Nevertheless, sulfide electrolytes are highly sensitive to moisture and oxygen, readily decomposing in air and releasing H_2S gas, which poses safety and cost challenges for material synthesis and large-scale production [9,15]. In addition, their interfacial stability with high-voltage cathode materials still requires further optimization [14].

In recent years, halide solid-state electrolytes (such as Li_3YCl_6 and Li_3InCl_6) have attracted growing interest because they combine relatively high ionic conductivity with good stability against high-voltage cathodes [5,18]. Studies indicate that these materials show promising compatibility with high-nickel cathode materials, although issues related to mechanical strength and cost remain to be addressed [18]. Furthermore, emerging inorganic systems such as glassy and glass-ceramic electrolytes continue to be reported, providing new avenues for expanding the solid-state electrolyte material library [11].

Polymer solid-state electrolytes (Polymer Solid Electrolytes, PSEs) typically employ polymer matrices such as poly(ethylene oxide) (PEO), in which lithium-ion transport is achieved through the dissolution of lithium salts [19]. These electrolytes offer advantages including good flexibility, excellent film-forming ability, and simple processing, giving them inherent benefits in interfacial contact and scalable manufacturing [2]. However, conventional polymer solid-state electrolytes generally exhibit low room-temperature ionic conductivity (typically below 10^{-5} $S \cdot cm^{-1}$), which limits their application in high-power devices [19]. Matthew Li et al. pointed out that the development of polymer electrolytes should move beyond single-material optimization and instead focus on systematic innovation through structural, interfacial, and functional co-design to adapt to the complex interfacial environments of solid-state batteries [20].

Composite solid-state electrolytes aim to simultaneously achieve high ionic conductivity and good mechanical flexibility by combining inorganic solid electrolytes with polymer matrices, and have become one of the most active research directions in recent years [8,18]. Studies have shown that inorganic fillers can not only serve as fast ion-conduction pathways, but also suppress polymer crystallization, improve mechanical properties, and enhance interfacial stability [18]. Depending on structural configuration, composite solid electrolytes can be classified into filler-dispersed types, continuous inorganic framework types, and layered composite structures. Although composite systems have demonstrated promising performance at the laboratory scale, they still face significant challenges in terms of interfacial uniformity, long-term stability, and scalable fabrication [9,27].

Overall, each class of solid-state electrolyte material system possesses distinct advantages and inherent limitations. Inorganic solid electrolytes exhibit clear superiority in ionic conductivity and safety, but suffer from processing and interfacial challenges; polymer systems are easy to process but constrained by performance limitations; composite systems offer potential for synergistic performance enhancement but involve higher engineering complexity [6,8,19]. Li Hong and Chen Liquan emphasized that future development of solid-state electrolytes should focus on the synergistic optimization of material system selection and interfacial engineering, and on establishing diversified technological routes tailored to different application scenarios, rather than seeking a single "optimal" material system [1]. This perspective provides important guidance for subsequent research on solid-state electrolyte manufacturing processes and interfacial engineering.

3 KEY PRODUCTION PROCESSES OF SOLID-STATE ELECTROLYTES AND RESEARCH PROGRESS

In addition to the challenges associated with material design, the preparation and processing technologies of solid-state electrolytes constitute critical factors that constrain the performance and industrialization of all-solid-state batteries. Different material systems exhibit significant differences in preparation conditions, process parameters, and production environments, and their processing routes directly influence the microstructure, ion transport behavior, and interfacial stability of solid-state electrolytes [6,9]. Therefore, a systematic review of key production processes and recent research progress is of great importance for translating material-level properties into device-level performance.

The solid-state reaction method is the most commonly used conventional route for preparing oxide solid-state electrolytes, typically involving raw material mixing, ball milling, pellet pressing, and high-temperature sintering [11-

12]. Garnet-type oxide electrolytes represented by LLZO usually require sintering at temperatures ranging from 1000 to 1200 °C to achieve high densification and stable crystal phases [13]. Studies have shown that sintering temperature, holding time, and atmospheric conditions significantly affect grain boundary structures and ionic conductivity. Although high-temperature sintering is beneficial for densification, it may also cause lithium volatilization, secondary phase formation, and increased grain boundary resistance [15]. Consequently, reducing sintering temperature through dopant regulation, the introduction of sintering aids, and atmosphere optimization has become an important research direction [6,13].

To address the high energy consumption and process complexity associated with traditional high-temperature sintering, mechanochemical synthesis methods, such as high-energy ball milling, have been applied to the preparation of sulfide and some oxide solid-state electrolytes [17]. This approach induces solid-state reactions through mechanical energy, enabling material synthesis at relatively low temperatures or even at room temperature, thereby significantly reducing energy consumption and shortening processing routes [9,17].

For sulfide solid-state electrolytes, mechanical milling combined with low-temperature annealing has become the dominant laboratory-scale preparation method, yielding amorphous or microcrystalline materials with high ionic conductivity [17]. However, this process imposes stringent requirements on powder homogeneity, equipment sealing, and post-treatment consistency, and still faces challenges during scale-up for industrial production [15].

The melt–quenching method is mainly employed for the fabrication of glassy and glass-ceramic solid-state electrolytes. In this process, raw materials are melted at high temperatures and rapidly cooled to form amorphous structures, followed by controlled crystallization to construct fast ion-conduction pathways [11]. Glass-ceramic electrolytes exhibit good ionic conductivity and structural uniformity; however, their high processing temperatures, strict equipment requirements, and limited compatibility with existing battery manufacturing processes restrict their large-scale application [6].

Polymer solid-state electrolytes are typically prepared using solution casting or melt extrusion techniques. Solution-based methods offer advantages such as uniform film formation and controllable thickness, but may introduce solvent residue and environmental burdens [19]. In contrast, melt extrusion is more compatible with continuous industrial manufacturing processes [2]. Studies have shown that regulating polymer molecular structures, lithium salt concentrations, and processing temperatures can partially improve the ionic conductivity and mechanical properties of polymer electrolytes [18-19].

The fabrication of composite solid-state electrolytes is more complex, generally involving the dispersion of inorganic fillers, interfacial modification, and the construction of multiphase architectures [8]. Common approaches include in situ polymerization, solution blending, and layered composite assembly [16]. Research indicates that the particle size, morphology, and interfacial chemistry of inorganic fillers play decisive roles in determining ion transport pathways. Improper filler distribution may lead to disrupted ionic conduction networks or increased interfacial resistance [9]. Therefore, achieving precise control over inorganic–organic interfaces remains a central challenge in composite electrolyte processing [18,20].

To reduce internal resistance and enhance energy density in solid-state batteries, the fabrication of ultrathin solid-state electrolyte layers has become an important research focus. Common thin-film preparation techniques include magnetron sputtering, pulsed laser deposition (PLD), and chemical vapor deposition (CVD) [10]. Thin-film electrolytes offer advantages such as precise thickness control and high densification, making them suitable for microbatteries and high-end devices [10]. However, their high cost and low throughput currently limit these techniques primarily to laboratory research and small-scale applications [6].

Extensive studies have demonstrated that the production processes of solid-state electrolytes influence not only their bulk ionic conductivity, but also profoundly determine the structure and stability of electrode/electrolyte interfaces [14,16]. For example, high-temperature sintering may promote the growth of interfacial reaction layers, whereas low-temperature forming may introduce porosity and poor interfacial contact [15]. Wang Han et al. emphasized that interfacial issues largely originate from microstructural mismatches introduced during material preparation and processing, highlighting the necessity of addressing interfacial challenges from the perspective of process optimization [16]. This viewpoint underscores the foundational role of production processes in interfacial engineering.

From an engineering perspective, current production processes for solid-state electrolytes still face several major challenges: (1) high-performance materials often require complex or costly processing routes; (2) insufficient consistency during scale-up from laboratory to industrial production; and (3) high sensitivity of preparation processes to environmental factors such as moisture and oxygen, which increases manufacturing difficulty [6,9,15].

4 INTERFACIAL ENGINEERING IN SOLID-STATE BATTERIES: CHALLENGES AND INNOVATIVE STRATEGIES

In all-solid-state battery systems, although significant progress has been achieved in the bulk ionic conductivity of solid-state electrolytes, their electrochemical performance in practical devices remains far below theoretical expectations. Extensive studies have demonstrated that interfacial issues, rather than bulk properties, constitute the primary bottleneck limiting performance enhancement and engineering application of solid-state batteries [14,16]. The ubiquitous solid–solid interfaces in solid-state batteries exhibit high complexity in structural, chemical, and mechanical aspects, rendering interfacial engineering one of the most critical and challenging research topics.

All-solid-state batteries mainly involve the following key interfaces: (1) solid-state electrolyte/cathode interfaces; (2)

solid-state electrolyte/anode interfaces, particularly those involving lithium metal; and (3) solid-state electrolyte/current collector interfaces [6,14].

Unlike the liquid–solid interfaces in conventional liquid electrolyte systems, the solid–solid interfaces in solid-state batteries generally suffer from incomplete contact and limited real contact area, which restricts ion transport pathways and results in significant interfacial resistance [16]. In addition, mismatches in crystal structure, chemical potential, and mechanical properties between different materials can induce stress concentration and structural degradation at interfaces [9].

Interfacial resistance is a key factor affecting the rate capability and cycle life of solid-state batteries. Studies have shown that space-charge layers (SCLs) are prone to form at the electrolyte/electrode interfaces, leading to local lithium-ion concentration distortion and suppressed cross-interface ion migration [14,16]. The origins of interfacial resistance include not only poor physical contact, but also changes in electronic structure, increased ion migration energy barriers, and localized interfacial chemical reactions [16]. Under high-voltage cathode conditions, electrochemical instability at interfaces further exacerbates resistance growth [21]. During battery operation, a series of unfavorable interfacial chemical reactions may occur between solid-state electrolytes and electrode materials, such as electrolyte decomposition and the growth of interphase layers [6,14]. These reactions consume active lithium, significantly increase interfacial resistance, and deteriorate cycling stability.

Taking sulfide solid-state electrolytes as an example, oxidative decomposition readily occurs at their interfaces with high-voltage cathode materials (such as high-nickel layered oxides), resulting in the formation of electronically insulating interfacial layers [14,21]. Although oxide solid-state electrolytes exhibit higher thermodynamic stability, interfacial reduction reactions and lithium dendrite penetration may still occur when they are in contact with lithium metal anodes [11,16].

Lithium metal anodes are regarded as an ideal choice for achieving ultra-high-energy-density solid-state batteries; however, their interfacial stability with solid-state electrolytes is particularly problematic [1,14]. Studies have revealed that even in solid electrolytes with high mechanical modulus, lithium dendrites may propagate along grain boundaries or defect regions, ultimately leading to internal short circuits [16].

Furthermore, volume changes and stress evolution at interfaces can induce contact failure and interfacial delamination, further degrading battery performance [9]. Therefore, suppressing lithium dendrite growth while maintaining high ionic conductivity represents one of the central scientific challenges in interfacial engineering [14].

Introducing ultrathin interfacial coatings (such as oxide, phosphate, or polymer layers) on electrode or solid-state electrolyte surfaces has been proven to effectively isolate detrimental interfacial chemical reactions and improve interfacial contact [6,16]. This strategy can significantly reduce interfacial resistance and enhance cycling stability [21].

Adjusting the chemical composition of solid-state electrolytes or constructing gradient interfacial structures enables gradual transitions in chemical potential and crystal structure across interfaces, thereby mitigating space-charge layer effects and improving interfacial stability [14,16]. Additionally, introducing flexible interlayers or designing composite interfacial architectures has been reported to effectively alleviate stress concentration at interfaces [9,18].

Overall, interfacial engineering has become the critical bridge linking the intrinsic material properties of solid-state electrolytes with the device-level performance of all-solid-state batteries. Current research has gradually shifted from passive remediation of single interfacial issues toward proactive design strategies involving the synergistic optimization of materials, structures, and processing technologies [6,16].

5 INDUSTRIALIZATION STATUS, TECHNOLOGICAL PATHWAYS, AND FUTURE PERSPECTIVES

The transition of solid-state batteries from laboratory research to large-scale application still faces multifaceted challenges related to materials, processes, and manufacturing systems [1,6]. This section analyzes the technological pathways and industrial development status in China, Japan, and Europe, and discusses future prospects.

China has adopted a pragmatic and diversified technological strategy, with sulfide-based electrolytes as the primary focus, while simultaneously advancing oxide and polymer-based routes. At the national level, large-scale industry–academia–research collaboration platforms have been established under the leadership of top academic teams, achieving breakthroughs in key materials and processes. For example, in situ solidification technologies have significantly reduced production costs. Industrialization has entered the process validation stage, with leading battery manufacturers such as CATL and BYD establishing pilot production lines. Companies including WeLion and QingTao have achieved mass production and vehicle installation of semi-solid-state batteries, and plan to realize large-scale application of all-solid-state batteries around 2027–2028. The primary challenges currently lie in resolving solid–solid interfacial stability, ensuring large-scale production consistency of sulfide electrolytes, and addressing high costs.

Japan has long been deeply engaged in the development of sulfide solid-state electrolytes and has built strong patent barriers. With robust government support, close alliances have formed between automakers (such as Toyota and Nissan) and materials companies (e.g., Idemitsu Kosan), with more than two decades of continuous research and development. Japan's industrialization roadmap is cautious yet clear, generally targeting the 2027–2030 timeframe as the commercialization window for all-solid-state batteries. For instance, Toyota plans to launch electric vehicles with driving ranges exceeding 1000 km during this period. Japan's core strengths lie in fundamental materials research and long-term accumulation, while the extremely high cost of sulfide-based systems and relatively slow industrialization pace present challenges for global market competition.

The development model in Europe and the United States is characterized by diversification and capital-driven innovation. Technological routes mainly focus on polymer and oxide systems, while sulfide-based electrolytes continue to be explored. Innovation momentum largely originates from start-ups such as QuantumScape and Solid Power, which are often spun out from leading academic research and supported by substantial venture capital, as well as strategic partnerships with major automakers including Volkswagen and BMW. The Western development strategy emphasizes achieving breakthroughs through disruptive technologies and has now entered a critical stage transitioning from laboratory prototypes to engineering-scale vehicle testing. However, a relatively weaker domestic large-scale lithium battery manufacturing ecosystem remains a key limitation for achieving mass production and cost control.

Overall, the global solid-state battery industry is currently in a transitional phase from laboratory prototypes to pilot-scale production. Existing solid-state battery products are mainly limited to small capacities, low-rate performance, or specific application scenarios, and a certain technological gap remains before large-scale commercialization.

From the perspective of technological pathways, sulfide-based electrolytes are widely regarded as one of the most promising routes for commercialization due to their high ionic conductivity and favorable interfacial contact [17]. This pathway facilitates low interfacial resistance and high rate capability, but imposes stringent requirements on production environments and entails high material and equipment costs [9,15]. Oxide-based routes offer superior chemical and thermal stability and thus advantages in safety [11]. However, their high-temperature sintering processes and relatively large interfacial resistance limit their application in high-energy-density batteries, necessitating complex interfacial engineering and structural design [14,16]. Polymer and composite solid-state electrolytes, owing to their relatively mature processing technologies, are considered important transitional routes from liquid to all-solid-state batteries [18-19]. While these routes exhibit higher near-term engineering feasibility, their performance ceilings remain lower than those of fully inorganic solid-state systems [2,20,22].

In summary, the core challenges facing the industrialization of solid-state batteries include: (1) the trade-off between high material performance and manufacturability of solid-state electrolytes; (2) interfacial resistance and long-term stability at electrode/electrolyte interfaces; (3) incomplete compatibility between existing lithium-ion battery manufacturing infrastructures and solid-state battery processes; and (4) cost control and consistency during large-scale production [1,6,15].

Looking ahead, future development of solid-state electrolytes and solid-state batteries is expected to exhibit several key trends. First, material design strategies will increasingly prioritize interfacial stability, with interfacial issues addressed at the early stages of material and structural design [16,20]. Second, process innovation oriented toward industrialization—including low-temperature sintering, continuous manufacturing, and compatibility with existing lithium battery equipment—will be crucial for accelerating commercialization [1,6]. Over an extended period, semi-solid, composite solid-state, and all-solid-state battery technologies are likely to coexist and develop in parallel to meet the diverse performance and cost requirements of different application scenarios [2,19].

6 CONCLUSION

This paper provides a systematic review of the research progress on solid-state electrolytes and their production processes. It comprehensively summarizes solid-state electrolyte material systems, key preparation technologies, interfacial engineering challenges, and the industrialization pathways of solid-state batteries, thereby establishing an integrated analytical framework spanning from fundamental materials research to engineering applications and industrial practice.

Future research should focus on forming a synergistic mechanism among high-performance material development, in-depth interfacial engineering, and manufacturing process innovation. Through coordinated advances in these aspects, solid-state batteries are expected to achieve substantial breakthroughs in safety, energy density, and cost control, providing strong support for the development of next-generation energy storage technologies.

COMPETING INTERESTS

The authors have no relevant financial or non-financial interests to disclose.

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