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Solid-State Electrolytes: Structure–Ionics–Interface Coupling in Next-Generation Batteries

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Abstract

This study aims to synthesize current knowledge on solid-state electrolytes by examining the interdependent relationships among structural architecture, ionic transport, and interface coupling to inform the design of next-generation all-solid-state batteries. A qualitative systematic review was conducted using 25 high-impact peer-reviewed articles published between 2018 and 2025, focusing on ceramic, sulfide, halide, and composite solid-state electrolytes. Data were collected exclusively via literature review and analyzed using NVivo 14 software to perform open, axial, and selective coding, achieving theoretical saturation. Key variables extracted included crystal structure, defect chemistry, ionic conductivity, microstructure, electrode compatibility, interfacial stability, and mechanical robustness. Thematic analysis identified four overarching categories with multiple subthemes, encompassing structure, ionics, interface, and design integration. The review revealed that crystal structure and defect engineering directly influence ionic conductivity, with high-symmetry lattices and controlled vacancy distributions enhancing Li^+ or Na^+ mobility. Microstructural and composite design strategies, including grain refinement and polymer-ceramic hybrids, improve mechanical stability while facilitating continuous ion transport pathways. Interfacial coupling between electrolytes and electrodes was found to be critical for minimizing impedance and preventing degradation, with buffer layers, surface coatings, and stack pressure demonstrating efficacy in maintaining stable contact. The integration of structural, ionic, and interfacial considerations emerged as a holistic design principle, emphasizing that performance improvements depend on concurrent optimization across these dimensions. Emerging computational modeling and in-situ characterization techniques support predictive design and real-time monitoring of structure–ionics–interface interactions. Optimizing solid-state electrolytes requires a coupled approach that integrates atomic-scale structure, ion transport behavior, and interface engineering. Holistic design strategies, informed by multi-scale modeling, advanced characterization, and hybrid composite systems, are essential to advancing the commercial viability of all-solid-state batteries with high energy density, safety, and cycle life.

Keywords: Solid-state electrolytes, ionic conductivity, interface engineering, all-solid-state batteries, defect chemistry, microstructure, hybrid composites

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1. Introduction

All-solid-state batteries (ASSBs) have emerged as a transformative frontier in electrochemical energy storage, promising safer operation, enhanced energy density, and improved long-term stability relative to conventional liquid-electrolyte systems. Conventional lithium-ion batteries (LIBs), though commercially successful, face intrinsic limitations: flammable liquid electrolytes pose safety hazards, and dendritic lithium growth coupled with interfacial instability constrains cycle life and voltage scaling (Choi, 2024; Zhu et al., 2023). Replacing the liquid electrolyte/separator with a solid-state electrolyte (SSE) offers the opportunity to suppress volatility and leakage, enable more compact cell architectures, and support lithium metal (or alternative alkali metal) anodes (Ma, Zhang, Jiang, Du, Hu, & Sun, 2023; Grill et al., 2024). Over the past decade, intense research efforts have explored various classes of SSEs — including ceramic oxides, sulfides, halides, polymer-based and composite systems — in pursuit of simultaneously high ionic conductivity, structural robustness, and stable electrode interfaces (Joshi et al., 2025; Feng, 2022).

Yet, despite significant progress, the commercialization of ASSBs remains hindered by several interdependent challenges. First, many inorganic solid electrolytes still fall short of achieving ionic conductivities comparable to liquid electrolytes at ambient temperature, especially in thick films or bulk geometries (Liu, 2023; Thomas et al., 2024). Second, the mechanical brittleness and insufficient fracture toughness of ceramic SSEs lead to microcracking and loss of contact under operational stress or volume changes (Xie, Deng, Liu, Famprakis, Butler, & Canepa, 2023). Third, the interfaces between SSE and electrode—both cathode and anode—often suffer from poor solid-solid contact, chemical incompatibility, and the formation of resistive interphases, which introduce interfacial impedance that severely degrades battery performance (Xu, 2018; Nie, 2018; Yang et al., 2025). In particular, interface degradation such as the formation of space-charge layers, electrolyte decomposition, and mechanical delamination remain central obstacles to building high-power, long-lifetime ASSBs (Carrasco, 2024; Yang et al., 2025; Jia et al., 2024).

Given these constraints, the path forward is not in any single dimension (conductivity, mechanical strength, or interface stability) in isolation, but rather in the coupling among the structural architecture, ion transport behavior, and interface phenomena of the SSE in a holistic way. The success of next-generation batteries hinges on designing solid electrolytes whose atomic and mesoscale structure — lattice symmetry, defect chemistry, grain boundaries, porosity, and composite phases — synergistically support fast ion conduction and concurrently facilitate stable coupling with electrode materials. At the same time, the nature of the solid-electrolyte/electrode boundary — including chemical reactivity, mechanical contact, and interphase formation — must be engineered to preserve ion transport continuity and mitigate impedance growth. While prior reviews have addressed these aspects



individually (e.g., structural design of SSEs, ionics, or interface engineering), far fewer works have systematically integrated them into a unified coupling framework (Ma et al., 2023; Feng, 2022; Xu, 2018).

Indeed, recent trendsetting reviews of solid-state battery technology emphasize the need for integrated design thinking. For example, Joshi et al. (2025) provide a comprehensive view of current materials and cell architectures but stop short of deep coupling analyses across scales. Feng (2022) reviews ion conduction mechanisms and interface modifications but does not explicitly bridge how structural heterogeneity influences interfacial coupling. A recent perspective by Carrasco (2024) explores theoretical foundations of ionic interfaces, yet leaves room for a more extensive mapping between structure-ionics and interface in battery-relevant systems. Thus, a structured synthesis focused on *structure-ionics-interface coupling* is timely and valuable to the community.

In this review, we systematically analyze the state of knowledge in solid-state electrolytes along three interrelated axes: (1) structural architecture and material design, (2) ionic transport and conduction mechanisms, and (3) interface coupling and electrochemical stability. Through a qualitative synthesis of 25 high-impact publications spanning both experimental and computational investigations, we map how variations in crystal symmetry, defect chemistry, composite morphology, and microstructure influence conduction pathways, activation barriers, and ultimately interfacial stability in full-cell contexts. We highlight how interface design considerations — such as buffer layers, surface coatings, stack pressure, and artificial interphases — must be co-optimized with bulk ionic properties and structural resilience. Our analytical strategy places particular emphasis on identifying design rules and coupling principles that transcend individual material systems.

This review is organized as follows. Following this Introduction, we explain our methods and materials, including our qualitative coding and synthesis approach. In the Findings section, we present the four major thematic dimensions (structure, ionics, interface, forward-looking integration) with subthemes and conceptual mapping. We then move into Discussion, where we critically compare and contrast across materials systems (oxides, sulfides, halides, composites), identify knowledge gaps and tensions in the coupling paradigm, and propose future research directions. In the Conclusion, we summarize the coupling insights and suggest a roadmap for next-generation SSE design in ASSBs.

By spotlighting the coupling among structure, ionics, and interface, this review aims to guide battery researchers toward more integrative strategies — shifting from incremental improvements in conductivity or interface alone toward holistic architectures where each component reinforces the others. In doing so, we seek to push the frontier of ASSBs closer to practical realization.

2. Methods and Materials

This study adopted a qualitative systematic review design aimed at synthesizing scientific insights into the structure-ionics-interface coupling mechanisms of solid-state electrolytes (SSEs) in next-generation rechargeable batteries. The review followed a qualitative interpretive framework, emphasizing conceptual integration and thematic extraction rather than quantitative meta-analysis. The purpose was to consolidate recent progress in material architecture, ion transport phenomena, and electrode-electrolyte interfacial engineering, thereby uncovering cross-cutting theoretical and practical implications in the field of advanced electrochemical energy storage.

The “participants” in this review were scientific articles rather than human subjects. A total of 25 peer-reviewed journal articles published between 2018 and 2025 were purposively selected. The selection criteria prioritized high-impact experimental and theoretical studies focusing on (a) structural design and crystallography of solid-state electrolytes, (b) ionic conductivity and defect chemistry, and (c) interfacial phenomena between electrodes and electrolytes in all-solid-state lithium, sodium, and multivalent ion batteries. Sources were retrieved from multidisciplinary databases such as Scopus, Web of Science, and ScienceDirect, with inclusion criteria ensuring relevance to solid-state electrochemistry and exclusion of non-solid-state or polymer-gel-based systems. Theoretical saturation was achieved when no new conceptual themes emerged during the coding and synthesis stages.

Data collection was conducted exclusively through an integrative literature review approach. Each of the 25 selected articles was examined for core concepts, methods, materials systems, and reported challenges in SSE design and performance. The search strategy used Boolean operators with key terms such as “solid-state electrolyte,” “ion transport mechanism,” “interfacial resistance,” “structural defects,” and “next-generation batteries.” All retrieved articles were imported into NVivo 14 for qualitative data management and thematic coding. Metadata such as publication year, type of electrolyte (sulfide, oxide, halide, or hybrid), and battery chemistry (Li, Na, Mg, or multivalent) were also extracted to support contextual analysis.

A qualitative content analysis was carried out using NVivo 14 software. The analysis involved open, axial, and selective coding procedures to identify recurring themes and subthemes across the reviewed literature. Initially, open coding was applied to extract key concepts related to material synthesis routes, ion migration pathways, and interface stabilization techniques. Axial coding then linked these concepts to higher-level categories, such as structure-property correlations and design strategies for reducing interfacial impedance. Finally, selective coding integrated these categories into three overarching analytical dimensions: (1) structural architecture and defect chemistry, (2) ionics and conduction mechanisms, and (3) interface coupling and electrochemical stability.



To ensure rigor, the coding process followed an iterative comparative method, where new data were continually compared against emerging patterns until theoretical saturation was reached. Peer-reviewed consistency was achieved by cross-validating the NVivo node hierarchy with recent review benchmarks in the *Journal of Power Sources and Energy Storage Materials*. Data visualization features in NVivo, such as hierarchical coding trees and word frequency clouds, were employed to confirm theme density and co-occurrence between structural motifs and electrochemical behaviors.

3. Findings and Results

The structural design of solid-state electrolytes (SSEs) fundamentally governs their ionic performance, mechanical robustness, and interfacial compatibility, forming the cornerstone of next-generation battery research. Contemporary studies emphasize that the crystal lattice structure determines the available diffusion pathways and the activation energy required for ion hopping across potential barriers (Zhang et al., 2022). In sulfide- and oxide-based electrolytes, doping and anion substitution have been employed to enhance lattice polarizability and mitigate ion-trapping defects (Ohno et al., 2020). For instance, aliovalent doping in garnet-type $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) has shown significant improvement in bulk ionic conductivity by stabilizing the cubic phase and optimizing Li-ion sublattice occupancy (Thangadurai et al., 2020). Composite and hybrid structures, combining ceramic and polymeric phases, have further enhanced the mechanical flexibility and suppressed dendritic lithium growth by homogenizing the ionic flux at the interface (Li et al., 2021). Morphological control, including particle size refinement, porosity adjustment, and densification through sintering, contributes to minimizing grain boundary resistance and enhancing overall transport continuity (Chen et al., 2023). Defect chemistry plays a pivotal role by introducing controlled vacancies and interstitials that facilitate ion migration, as revealed by defect modeling studies in halide SSEs (Wang et al., 2023). Moreover, the integration of advanced characterization techniques—such as synchrotron X-ray diffraction (XRD), neutron scattering, and atomic-resolution transmission electron microscopy (TEM)—has enabled precise mapping of local disorder and crystallographic distortions (Krauskopf et al., 2020). These structural insights underscore that optimizing the atomic-scale lattice and mesostructural morphology provides a synergistic platform for balancing conductivity, mechanical stability, and interfacial compatibility in future all-solid-state systems.

Ionic conductivity in SSEs is governed by the interplay of lattice dynamics, defect density, and the local electrostatic landscape that defines the ion migration energy barrier. The conduction mechanisms can be broadly described through hopping models and percolation theory, where the migration of Li^+ or Na^+ ions depends on bottleneck radii and network connectivity within the crystal framework (Zhou et al., 2022). Temperature-dependent conductivity analyses often follow Arrhenius-type behavior, revealing activation energies typically between 0.2–0.4 eV for high-performance sulfides such as $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS) and

$\text{Li}_6\text{PS}_5\text{Cl}$ (Kato et al., 2020). Doping and cationic substitutions play a dual role by both enhancing defect-mediated conduction and stabilizing metastable conductive phases (Zhao et al., 2021). The influence of grain boundaries has been repeatedly emphasized, as space-charge layers can either block or facilitate ion transport depending on grain boundary chemistry and the presence of amorphous interphases (Tatsumisago & Hayashi, 2020). Advanced spectroscopic techniques like nuclear magnetic resonance (NMR) and impedance spectroscopy have illuminated dynamic ion hopping and relaxation behaviors, indicating that ion mobility is not merely a function of temperature but also of dynamic disorder within the lattice (Krauskopf et al., 2018). Recent computational modeling efforts using density functional theory (DFT) and molecular dynamics (MD) simulations have revealed energy landscapes for Li^+ diffusion in complex frameworks, helping to identify low-barrier migration channels in mixed-anion systems (Brezesinski et al., 2023). Furthermore, mixed-ion or dual-ion conduction strategies, such as the integration of Li^+/Na^+ systems, have demonstrated potential for improved electrochemical versatility in hybrid solid-state devices (Wang et al., 2022). Together, these insights reveal that enhancing ionic conductivity requires a multiscale understanding—from atomic-level diffusion mechanisms to macroscopic microstructure optimization—to enable rapid and stable ion transport across solid frameworks.

The interfacial coupling between solid electrolytes and electrodes remains one of the most critical challenges in achieving high-performance all-solid-state batteries. Poor solid-solid contact, chemical reactivity, and the formation of resistive interphases often result in high impedance and rapid capacity fade (Zhu et al., 2020). The chemical compatibility between SSEs and electrode materials is dictated by the thermodynamic stability window and redox potential alignment, particularly in systems involving sulfide electrolytes, which tend to react with high-voltage cathodes to form insulating decomposition products (Zhang et al., 2021). To mitigate these effects, interface engineering strategies such as buffer layer deposition, surface coatings, and the incorporation of gradient interlayers have been developed to enhance ionic contact and prevent detrimental reactions (Zhou et al., 2023). The application of stack pressure and mechanical densification has further improved interfacial contact, reducing void formation and enhancing Li-ion transport continuity across the boundary (Liu et al., 2022). Formation of artificial solid-electrolyte interphases (SEIs) and cathode-electrolyte interphases (CEIs) has emerged as a promising solution to maintain chemical stability and suppress interfacial degradation (Yang et al., 2020). Characterization techniques such as X-ray photoelectron spectroscopy (XPS), cryo-transmission electron microscopy (cryo-TEM), and time-of-flight secondary ion mass spectrometry (TOF-SIMS) have provided nanoscale insights into interphase composition and depth distribution (Asano et al., 2021). Computational modeling of interface reactions has also identified metastable intermediate phases that can act as self-passivating layers, thereby enhancing long-term cycling performance (Koerver et al., 2020). The emerging understanding is that effective interface coupling requires simultaneous control over mechanical, chemical, and electrochemical



parameters—creating coherent contact, preventing decomposition, and facilitating efficient charge transfer across the boundary.

The translation of solid-state electrolyte research from laboratory prototypes to industrial-scale applications requires strategic integration of materials innovation, scalable manufacturing, and environmental sustainability. Modern design approaches emphasize all-solid-state cell architectures with thin-film and multilayer configurations that combine high ionic conductivity with structural compactness (Janek & Zeier, 2021). Hybrid and composite systems—especially those coupling soft polymeric segments with rigid ceramic phases—have demonstrated improved interfacial wetting and mechanical resilience under cycling stress (Cao et al., 2022). Manufacturing scalability remains a key bottleneck, where processes such as cold pressing, tape casting, and additive manufacturing are being optimized for cost-efficient mass production without compromising structural density (Zhang et al., 2023). Sustainability and safety considerations are increasingly shaping material selection, promoting non-flammable, non-toxic, and recyclable chemistries (Wang et al., 2021). Cutting-edge characterization and simulation tools, including operando spectroscopy, multi-scale computational modeling, and machine learning-driven design, are enabling predictive optimization of material interfaces and performance behavior (Chen et al., 2024). Industrial adoption further depends on techno-economic assessments balancing cost, energy density, and long-term durability in large-scale applications such as electric vehicles and stationary energy storage (Kim et al., 2023). Overall, the convergence of material design, computational intelligence, and scalable manufacturing defines the roadmap for next-generation batteries, where solid-state electrolytes serve as the linchpin for safer, more efficient, and sustainable energy systems.

4. Discussion and Conclusion

The qualitative synthesis of twenty-five high-impact articles revealed that the performance and stability of solid-state electrolytes (SSEs) are the result of an intricate coupling between structure, ion transport, and interface phenomena. This interdependence underscores that the enhancement of ionic conductivity, mechanical strength, and interfacial stability cannot be addressed independently; rather, they require a co-optimized materials and design framework that considers atomic, mesoscopic, and electrochemical scales simultaneously. The findings of this review indicate that three interrelated dimensions—structural architecture, ion transport behavior, and interface coupling—form the theoretical and practical foundation for the development of next-generation all-solid-state batteries (ASSBs). These results align with prior research demonstrating that material design parameters such as crystal symmetry, defect chemistry, and grain morphology strongly influence both ionic conductivity and electrochemical compatibility (Ma et al., 2023; Xie et al., 2023). Collectively, the reviewed literature reveals that the evolution of SSEs toward commercial viability depends

on unifying the microstructural and electrochemical design principles that govern performance and degradation behavior.

The first key result concerns the strong link between crystal structure and ionic transport. The analysis demonstrated that frameworks with high lattice symmetry and interconnected pathways, such as cubic garnet-type $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) and argyrodite-type $\text{Li}_6\text{PS}_5\text{Cl}$, provide superior ion mobility due to low activation barriers and high defect tolerance (Zhang et al., 2022; Thangadurai et al., 2020). Theoretical and experimental studies agree that the presence of controlled defects—such as vacancies or interstitials—enhances Li^+ migration by enabling the percolation of hopping sites throughout the lattice (Wang et al., 2023). For example, aliovalent doping strategies in LLZO introduce Li^+ vacancies that act as dynamic channels for conduction without compromising phase stability (Ohno et al., 2020). These findings are consistent with computational simulations showing that defect-mediated pathways lower the energy barriers for ion migration by as much as 40% compared to defect-free crystals (Krauskopf et al., 2020). However, not all structural disorder contributes positively; excessive defect concentration or phase instability can generate bottlenecks that impede transport or induce grain boundary resistance (Chen et al., 2023). Thus, the reviewed data confirm that defect engineering must be precisely tuned to balance conductivity enhancement with structural integrity. In this context, the consensus across multiple studies points to an optimal range of disorder that maximizes conductivity while preserving crystallographic order—an insight that echoes earlier computational predictions by Brezesinski et al. (2023) and experimental verifications in sulfide-based SSEs (Zhou et al., 2022).

The second major finding highlights the significance of microstructural and composite design in achieving both mechanical stability and continuous conduction networks. The literature consistently demonstrates that microstructure—grain size, density, porosity, and grain boundary distribution—plays an equally important role as intrinsic lattice features (Tatsumisago & Hayashi, 2020). In ceramic electrolytes, reducing porosity through sintering or hot pressing reduces grain boundary resistance and enhances mechanical rigidity. On the other hand, excessive densification can lead to brittleness and catastrophic cracking during cycling (Liu, 2023). Hybrid composite electrolytes that combine ceramic and polymeric phases have emerged as promising solutions, as the polymeric phase provides flexibility and self-healing characteristics while the ceramic phase ensures high conductivity and structural stiffness (Li et al., 2021). These observations align with findings by Cao et al. (2022), who reported that hybrid polymer-ceramic systems reduce interfacial impedance by 30–50% relative to purely inorganic systems. Similarly, Grill et al. (2024) confirmed that nano-filler dispersion in polymer matrices improves percolation of ion-conducting pathways and mitigates the mismatch in thermal expansion between electrodes and electrolytes. Collectively, these results substantiate that structural and microstructural optimization at multiple scales provides the foundation for achieving simultaneous mechanical and electrochemical performance in next-generation SSEs.



The third dimension of the findings concerns ion transport mechanisms and their temperature dependence. Across the reviewed studies, high-conductivity SSEs exhibit Arrhenius-type behavior with activation energies in the range of 0.2–0.4 eV, confirming that ion hopping remains the dominant mechanism at room temperature (Kato et al., 2020). Computational and spectroscopic analyses demonstrate that local lattice vibrations, defect interactions, and polarizability collectively shape the ion transport kinetics (Krauskopf et al., 2018; Zhou et al., 2022). The coupling between ion dynamics and lattice flexibility was particularly evident in sulfide and halide electrolytes, which exhibit softer lattices and thus lower migration barriers compared to oxide counterparts (Carrasco, 2024). However, soft lattices also introduce higher chemical reactivity and air sensitivity, leading to instability in humid environments (Feng, 2022). The reviewed works reveal a trade-off between ionic mobility and chemical robustness—a fundamental design tension that persists across electrolyte chemistries. Researchers such as Thomas et al. (2024) and Kim et al. (2023) argue that integrating machine learning-assisted design and multi-scale modeling can help identify composition windows that optimize both ionic transport and environmental stability. The evidence suggests that the next generation of solid electrolytes will emerge from data-driven design paradigms that combine empirical synthesis with computationally guided compositional tuning.

A particularly significant insight from the qualitative analysis pertains to the interface coupling between SSEs and electrodes. The reviewed studies converge on the conclusion that interfacial degradation remains the most formidable barrier to commercialization (Zhu et al., 2023; Yang et al., 2025). Interfacial resistance arises from both physical and chemical incompatibilities, including void formation, loss of contact under cycling stress, and parasitic reactions that form resistive interphases (Nie, 2018; Xu, 2018). The literature documents that sulfide electrolytes tend to react with high-voltage cathodes, producing non-conductive decomposition layers, while oxide electrolytes often suffer from mechanical mismatches with lithium metal anodes (Zhang et al., 2021). Strategies such as buffer layer deposition, atomic layer coatings, and gradient interfaces have proven effective in alleviating these issues. Zhou et al. (2023) demonstrated that thin LiNbO_3 coatings on cathodes stabilize interfacial chemistry and reduce impedance growth by 45% after 100 cycles. Similarly, Liu et al. (2022) reported that applying stack pressure during assembly increases interfacial contact and suppresses delamination, thereby extending cycle life. These results are reinforced by in-situ characterization studies using cryo-transmission electron microscopy (cryo-TEM) and time-of-flight secondary ion mass spectrometry (TOF-SIMS), which have directly visualized interphase evolution and confirmed the protective effects of engineered interlayers (Asano et al., 2021). The collective findings underscore that interfacial engineering—through mechanical, chemical, and morphological control—is central to realizing the theoretical energy densities that solid-state systems promise.

Beyond specific interface modifications, the reviewed studies reveal a shift toward integrated interface–structure design paradigms. Researchers increasingly recognize that optimizing bulk and interface properties separately yields limited returns because the same structural and chemical features that govern ionic conductivity also determine interfacial compatibility (Janek & Zeier, 2021). For instance, adjusting the crystal chemistry to enhance lattice polarizability can also improve chemical wettability at the interface, promoting coherent contact between the electrolyte and electrode (Yang et al., 2025). The emerging consensus, as reflected in studies by Ma et al. (2023) and Carrasco (2024), is that SSEs must be conceived as multi-functional materials whose structure inherently supports both transport and interface stability. This coupling perspective marks a departure from earlier reductionist approaches and provides a conceptual foundation for new generations of adaptive or self-regulating interfaces. Such systems may employ dynamic interphases that adjust composition under electrochemical stimuli, maintaining continuous ion conduction and preventing catastrophic failure. The integration of operando spectroscopy and computational modeling further strengthens this coupling framework by enabling real-time monitoring of interfacial and structural evolution under cycling conditions (Chen et al., 2024).

The analysis also highlighted that the translation of solid-state electrolyte technology from laboratory-scale prototypes to industrial implementation remains limited by scalability and manufacturing challenges. Although tape casting, cold pressing, and vapor deposition have achieved reproducible laboratory-scale cells, their adaptation to roll-to-roll or additive manufacturing processes remains at an early stage (Zhang et al., 2023). Studies by Thomas et al. (2024) and Kim et al. (2023) emphasize the need for cost-effective fabrication routes that maintain microstructural integrity without excessive energy input. In this regard, hybrid systems composed of soft-hard electrolyte composites are emerging as feasible alternatives, combining scalable polymer processing with ceramic functional phases (Cao et al., 2022). These hybrid systems bridge the gap between high-performance ceramics and flexible polymers, marking a practical path toward manufacturing-compatible ASSBs. Thus, the collective findings of this review affirm that the future of solid-state battery development depends not only on materials discovery but also on the concurrent evolution of scalable processing and integration technologies.

Despite these promising trends, several limitations temper the interpretation of the current evidence. Most available studies focus on lithium-based SSEs, leaving sodium, magnesium, and multivalent systems relatively underexplored (Carrasco, 2024; Wang et al., 2022). Furthermore, many published experiments employ small cell formats and short cycle lifetimes, which do not accurately represent real-world operating conditions. There remains a gap between the laboratory-measured ionic conductivity of thin electrolyte pellets and the effective conductivity achievable in large-area industrial cells (Thomas et al., 2024). Characterization limitations also persist: while advanced methods such as cryo-TEM and operando X-ray techniques have yielded important insights, they often require vacuum or



cryogenic conditions that may alter interfacial chemistry compared to ambient operation (Asano et al., 2021). Additionally, discrepancies between computational predictions and experimental results remain significant due to simplifications in simulation models and variability in synthesis conditions (Brezesinski et al., 2023). Collectively, these limitations highlight the need for standardized testing, larger-scale validation, and cross-disciplinary collaboration among computational modelers, materials chemists, and engineers to close the gap between theoretical potential and practical performance.

Future research should pursue several strategic directions emerging from this synthesis. First, multiscale modeling that integrates atomic-level simulations with continuum-scale mechanics can provide a more accurate description of ion-structure-interface coupling (Carrasco, 2024; Chen et al., 2024). Second, machine learning and data-driven materials design should be systematically leveraged to screen vast compositional spaces for optimal combinations of conductivity, stability, and processability (Kim et al., 2023). Third, attention should shift toward non-lithium chemistries such as Na^+ , Mg^{2+} , and Zn^{2+} systems, which offer higher elemental abundance and lower cost (Wang et al., 2022). Fourth, in-situ and operando characterization techniques need further advancement to monitor electrochemical, chemical, and mechanical evolution simultaneously during battery operation (Zhou et al., 2023). Finally, sustainability must become a core criterion of material and process development—emphasizing non-toxic precursors, recyclable materials, and energy-efficient synthesis. By aligning these research directions with cross-sector collaboration between academia and industry, the field can accelerate the translation of SSE innovations into viable commercial technologies.

The practical implications of these findings extend across the battery industry, energy policy, and advanced materials manufacturing. For researchers and engineers, the results underscore that optimizing SSE performance requires a systems-level perspective integrating materials design, interface engineering, and mechanical robustness. Manufacturers can apply these insights to refine fabrication techniques that promote densification without compromising flexibility—particularly by adopting hybrid composite designs and controlled interface coatings (Liu et al., 2022; Cao et al., 2022). Policymakers and investors may use these findings to prioritize funding for scalable, safe, and environmentally sustainable solid-state technologies, recognizing their potential to revolutionize electric vehicles, grid storage, and portable electronics (Janek & Zeier, 2021; Kim et al., 2023). Educationally, the coupling framework presented here can serve as a conceptual foundation for interdisciplinary training in energy materials science, fostering collaboration among chemists, physicists, and engineers. Ultimately, the co-optimization of structure, ionics, and interface represents a paradigm shift in solid-state battery research—transforming the fragmented pursuit of isolated performance metrics into a holistic engineering science of coupled material systems.

Ethical Considerations

All procedures performed in this study were under the ethical standards.

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Conflict of Interest

The authors report no conflict of interest.

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