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Challenges and Design Strategies for Solid Electrolytes in All-Solid-State Batteries: A Bibliometric and Critical Review

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Abstract. All-solid-state batteries (ASSBs) have attracted extensive attention as a potential next-generation energy-storage technology because they may combine high energy density, improved safety, and compatibility with lithium-metal anodes. However, the replacement of liquid electrolytes with solid ion conductors introduces new limitations involving ionic transport, solid–solid interfaces, mechanical degradation, lithium dendrite growth, and scalable manufacturing. This review combines a bibliometric perspective with a critical discussion of these challenges and the associated design strategies for solid electrolytes. Based on the Web of Science dataset described in the research materials, the field has experienced rapid growth, particularly since 2020, while research activity has become concentrated in a number of major institutions and interconnected research groups. The review then examines sulfide, oxide, polymer, and composite electrolytes from the perspective of bulk transport, chemical stability, mechanical properties, and processability. Particular attention is given to interface engineering, artificial interphases, defect and crystal-structure regulation, and chemomechanical design. The comparison indicates that no single electrolyte family simultaneously satisfies all requirements for practical ASSBs. Future progress will therefore depend on integrated optimization of electrolyte chemistry, microstructure, interfaces, electrode architecture, manufacturing conditions, and cost. Standardized testing, advanced operando characterization, data-driven materials discovery, and scalable low-pressure manufacturing are expected to play increasingly important roles in translating laboratory demonstrations into commercially relevant solid-state batteries.

Keywords: All-solid-state batteries, Solid electrolytes, Lithium-metal anode, Solid-solid interface, Interface engineering

1. Introduction

The growing demand for safer, higher-energy-density rechargeable batteries has intensified interest in ASSBs. In contrast to conventional lithium-ion cells that use flammable organic liquid electrolytes, ASSBs employ solid ion conductors and can potentially be paired with lithium-metal anodes. These features offer a route toward improved safety and higher cell-level energy density, although the benefits depend strongly on electrolyte thickness, electrode loading, inactive-material content, and operating pressure [1-4]. Replacing the liquid phase therefore does not eliminate safety

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and reliability concerns; instead, it shifts the central problems toward solid-state transport, buried interfaces, mechanical damage, and manufacturability [4, 5].

A practical solid electrolyte must combine high Li-ion conductivity with low electronic conductivity, chemical and electrochemical compatibility, adequate fracture resistance, and scalable processability [1, 2, 6]. Sulfide electrolytes can reach room-temperature conductivities comparable to liquid electrolytes and can be densified at moderate pressure [7-9], but many are moisture-sensitive and react with electrode materials. Oxide electrolytes generally provide better thermal and chemical robustness, yet their rigidity and grain-boundary resistance make intimate solid–solid contact difficult [2, 6, 10]. Polymer electrolytes are flexible and processable, but segmental-motion-controlled transport often gives insufficient room-temperature conductivity [11]. Polymer–ceramic composites seek to combine inorganic transport pathways with polymer compliance [12]. At the cell level, interfacial reactions, void formation, lithium penetration, and chemomechanical degradation remain decisive limitations [4, 5, 13].

The rapid expansion of solid-state battery research makes it difficult to distinguish durable research directions from isolated demonstrations. Existing reviews cover electrolyte chemistry, lithium metal, interfaces, and practical cell design [1-6, 10, 13, 14]. This article therefore combines a bibliometric overview with a critical literature review. Publication trends and institutional contributions are first examined using the stated Web of Science dataset. The discussion then connects those patterns to the principal challenges in ion transport, interface stability, lithium-metal cycling, mechanical integrity, and manufacturing. The aim is to identify design strategies that improve complete-cell performance rather than only a single materials metric.

2. Bibliometric analysis

2.1. Data collection and bibliometric methods

To obtain an overall view of the field, a bibliometric analysis was conducted using the Web of Science (WoS) Core Collection. The dataset contains 36,881 solid-state-battery-related records, including titles, authors, affiliations, keywords, and citation information. Bibliometric analysis provides a reproducible way to examine the structure and evolution of a large research corpus, provided that the search query, timespan, document types, and data-cleaning rules are reported transparently [15, 16].

The analysis focuses on annual publication output, institutional contributions, and keyword co-occurrence. Publication counts were used to examine growth; affiliation fields were normalized before institutional ranking; and recurring author keywords were used to construct an overlay network. VOSviewer was used for network mapping because it supports large co-occurrence maps and overlay visualization [17]. The numerical results should be interpreted as properties of the exported dataset rather than as an exhaustive census of every solid-state-battery publication.

2.2. Research trends and hotspot analysis

As shown in Figure 1, the dataset exhibits sustained publication growth from 2015 to 2025, with a marked acceleration after 2020. This trend is consistent with the broader shift of solid-state batteries from electrolyte discovery toward lithium-metal integration, high-loading electrodes, and manufacturable cell architectures [18, 19].

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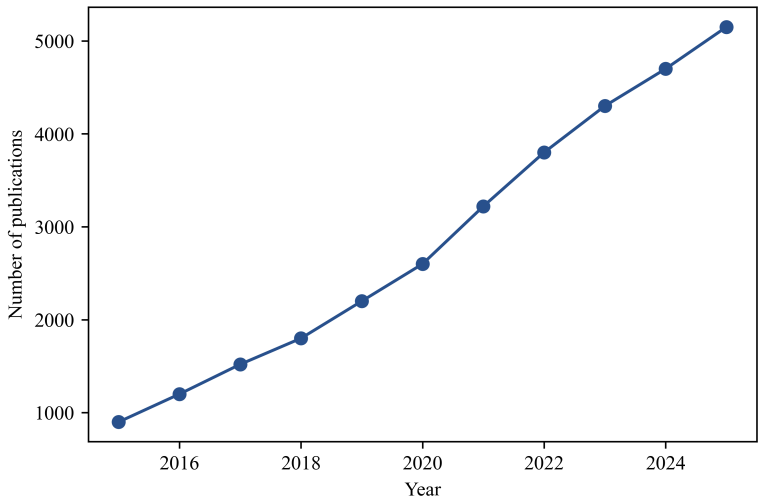


Figure 1. Publication trend of all-solid-state battery research based on Web of Science dataset

Figure 2 shows that publications in the dataset are concentrated within a small number of institutions, predominantly from China. Rankings may vary depending on how institutional name variants, affiliated organizations, and fractional counting are handled. Nevertheless, this distribution highlights the multidisciplinary and collaborative nature of ASSB research, which spans solid-state chemistry, electrochemistry, mechanics, characterization, and manufacturing.

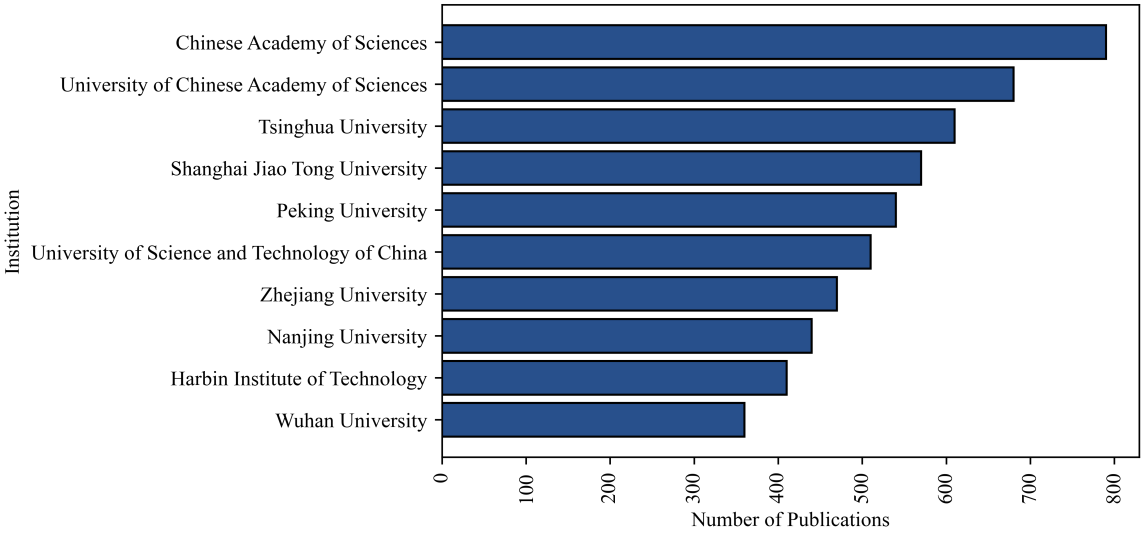


Figure 2. Top-ten contributing institutions for all-solid-state battery research based on Web of Science dataset

3. Challenges for solid-state batteries

3.1. Limited ionic conductivity of solid electrolytes

Fast Li-ion transport is a prerequisite for high-rate ASSBs. Sulfide conductors such as LGPS-related phases can reach room-temperature conductivities in the 10^{-2} S cm^{-1} range, but moisture sensitivity and interfacial instability complicate handling and operation [7-9]. Oxide electrolytes offer stronger

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thermal and chemical robustness, although grain boundaries, porosity, and poor contact can dominate their effective resistance [2, 6, 20]. Polymer electrolytes are flexible and readily processed into thin membranes, yet their conductivity falls sharply near room temperature when segmental chain motion is restricted [11]. Thus, conductivity measured for a bulk specimen is necessary but not sufficient for a practical cell.

Transport measured in dense pellets does not fully represent a composite electrode or thin separator in an operating cell. Tortuosity, particle packing, electrolyte thickness, active-material loading, grain boundaries, and contact area collectively determine the effective ion pathway.

3.2. Interfacial instability between electrodes and solid electrolytes

Even when a solid electrolyte has excellent bulk conductivity, stable cycling requires efficient Li-ion transfer across electrode–electrolyte interfaces. Unlike a liquid, a solid cannot spontaneously wet newly created surfaces. Assembly defects, roughness, and cycling-induced gaps therefore reduce the true contact area and constrict current, causing interfacial resistance to rise [5, 14].

Chemical and electrochemical incompatibility further aggravate interface degradation. Sulfide electrolytes can oxidize against high-voltage oxide cathodes, while several inorganic electrolytes are reduced by lithium metal; the resulting interphases may be passivating or may continue to grow, depending on their ionic and electronic transport properties [4, 5]. In composite cathodes, volume change and anisotropic strain can create microcracks and contact loss, while reaction layers impede charge transfer and contribute to capacity fade [21].

Interface engineering has consequently become a central ASSB design strategy. Thin oxide, phosphate, or Li-ion-conducting coatings can suppress direct reactions at cathode–electrolyte contacts, while artificial interlayers and composition gradients can improve wetting and reduce mechanical mismatch [5, 22]. These approaches are effective only when the added layer remains thin, ionically conductive, electronically blocking where required, and mechanically stable throughout cycling.

3.3. Lithium dendrite growth and mechanical failure

Lithium metal is attractive because of its high theoretical specific capacity and low electrochemical potential, but its behavior against a solid electrolyte is governed by local current distribution, interfacial contact, creep, fracture, and interphase chemistry [3, 23]. The use of a mechanically stiff separator alone is therefore not a guarantee against internal short circuit.

Lithium can penetrate inorganic electrolytes through surface flaws, pores, grain boundaries, and cracks when local electrochemical driving forces and stress exceed the tolerance of the electrolyte [24]. During stripping, voids form if lithium is removed faster than creep or diffusion can replenish the interface; subsequent plating then concentrates current near the remaining contacts and promotes filament growth [25]. These coupled void–crack–filament processes explain why critical current density depends strongly on surface condition, pressure, temperature, and test protocol [23, 25].

Figure 3 presents an original synthesis of the principal interfacial failure processes that operate concurrently in ASSBs: contact loss, void-assisted dendrite growth, lattice mismatch, interdiffusion, space-charge and Li immobilization, and thermal–mechanical damage [5, 14, 21, 23-25]. These processes reinforce one another through local current constriction, heat generation, stress concentration, and fracture. Mitigation therefore requires coordinated control of current density, interface chemistry, surface defects, pressure, and stress relaxation rather than a single mechanical-property target [23-25].

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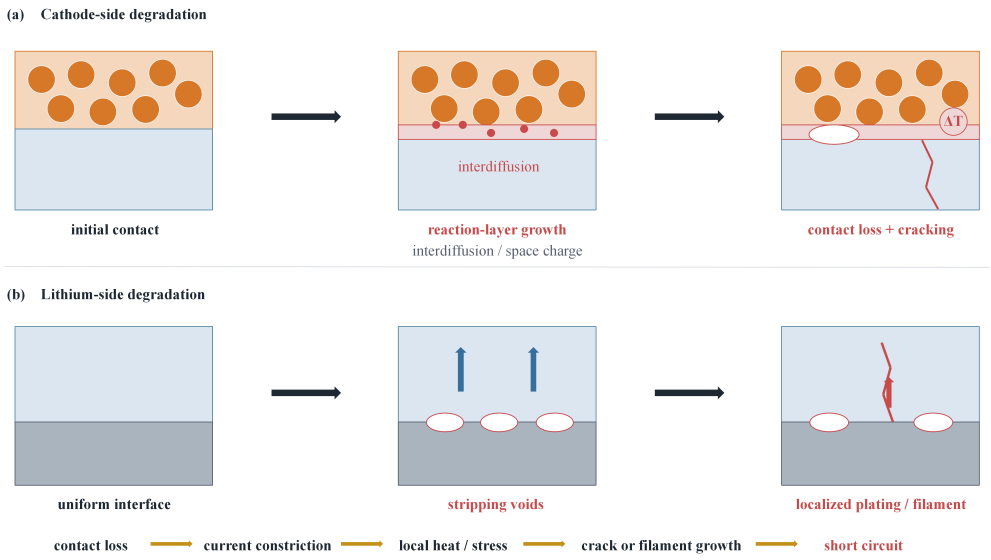


Figure 3. Coupled degradation pathways in all-solid-state batteries. (a) Cathode-side reaction-layer growth, interdiffusion, contact loss, and cracking. (b) Lithium-side stripping voids, current constriction, and localized filament growth

3.4. Challenges in practical battery manufacturing

Translating laboratory ASSBs into commercial products remains difficult because many high-performance demonstrations depend on excess lithium, thick electrolyte pellets, low active-material loading, small electrode area, elevated temperature, or high stack pressure. These conditions can inflate apparent performance while reducing practical specific energy and increasing pack complexity [19].

Practical cells require thin, defect-free electrolyte membranes and high-loading composite cathodes. Thinning the electrolyte improves energy density but increases sensitivity to pinholes, fracture, and thickness variation. Increasing cathode loading lengthens ion and electron pathways and magnifies reaction heterogeneity. Consequently, cell-level comparisons should report areal capacity, electrolyte thickness, pressure, temperature, and inactive-material fractions in addition to gravimetric capacity [19].

Manufacturing consistency is equally important. Powder handling, moisture control, mixing, densification, lamination, and interface formation all affect cell impedance and lifetime. Scalable processing must therefore be developed together with electrolyte chemistry, with particular attention to low-pressure operation, thin-film quality control, material cost, and compatibility with established roll-to-roll manufacturing [4, 19].

4. Solid electrolyte design strategies

4.1. Composition engineering

The electrochemical performance of a solid electrolyte is strongly governed by crystal structure, composition, and defect chemistry. Aliovalent substitution can change carrier concentration, site occupancy, phase stability, lattice polarizability, and migration barriers. Garnet LLZO provides a representative example in which composition and phase stabilization strongly affect Li-ion transport

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[26, 27], while the argyrodite family illustrates how anion chemistry and lattice dynamics regulate conductivity [28]. Figure 4 connects these composition-level variables to a primary LLZO study showing how Al site occupancy can block different fractions of the three-dimensional Li-conduction network [29]. High-voltage stability still requires coordinated intrinsic and extrinsic design [30].

Composition optimization often creates trade-offs. Dopants that enhance conductivity may narrow electrochemical stability, increase moisture sensitivity, introduce costly elements, or complicate synthesis. A useful composition must therefore be evaluated against conductivity, electronic leakage, stability, mechanical response, abundance, and compatibility with both electrodes, rather than being selected by a single record value [1, 2, 6].

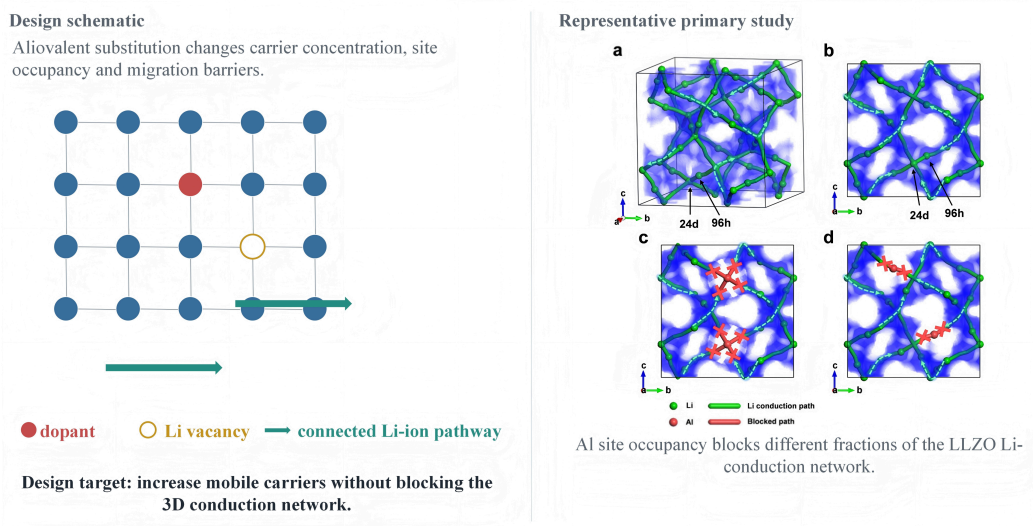


Figure 4. Composition engineering: aliovalent substitution, vacancy control, and site occupancy tune Li-ion migration pathways. The design schematic is paired with a primary-research example reproduced from Shin et al. [29], Fig. 4

4.2. Microstructure engineering

Microstructure controls how intrinsic transport translates into a device. Grain-boundary chemistry, residual porosity, particle morphology, texture, and densification affect both resistance and fracture behavior. In LLZO, different grain-boundary structures can contribute differently to Li transport and may also provide sites for current localization [20].

Processing should therefore aim for a uniform, sufficiently dense electrolyte while limiting secondary phases, lithium loss, and thermal damage. Conventional sintering, pressure-assisted routes, and low-temperature approaches each impose different trade-offs. Cold sintering illustrates how transient solvents and pressure can densify some ceramic systems at greatly reduced temperatures, although chemistry-specific validation is required before applying the method to a battery electrolyte [31]. Microstructural optimization should be linked to measured through-thickness conductivity, fracture resistance, and cell-level performance. Figure 5 illustrates this design logic and pairs it with FE-SEM evidence that a sintering aid changes the density, porosity, and grain-boundary structure of LGLZO [32].

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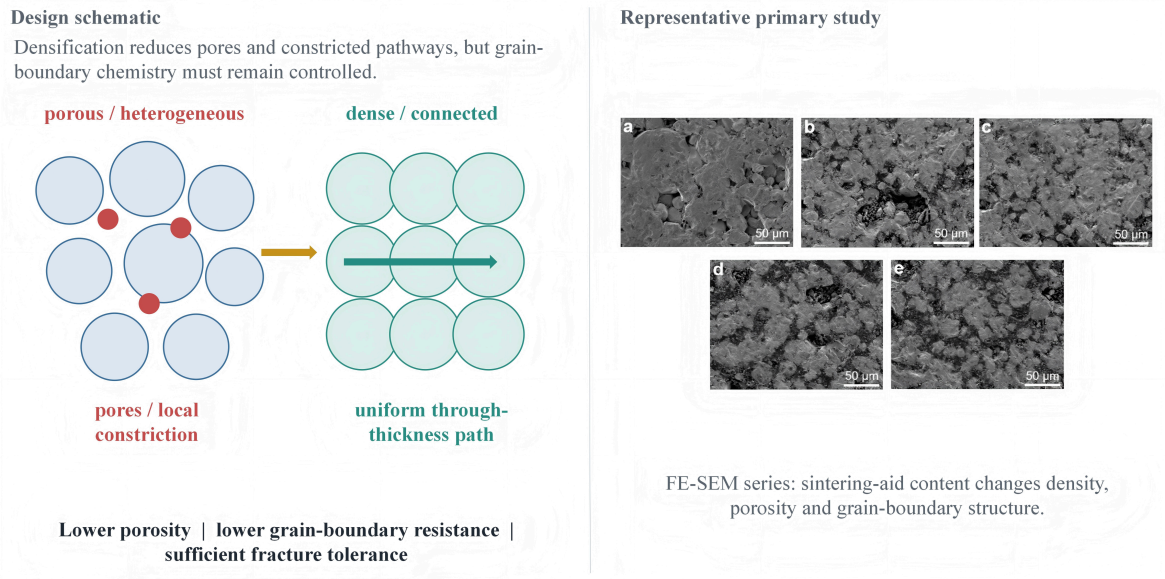


Figure 5. Microstructure engineering: densification and grain-boundary control convert intrinsic conductivity into effective through-thickness transport. The design schematic is paired with FE-SEM images reproduced from Lee et al. [32], Fig. 1

4.3. Interface engineering

Interface engineering addresses both physical contact and chemical stability. Its design variables include surface energy, interphase composition, ionic/electronic conductivity, elastic response, thickness, and processing compatibility. These factors are particularly important at lithium-metal and high-voltage-cathode interfaces, where reaction and stress evolve together [4, 5, 14, 21].

Cathode coatings based on oxides, phosphates, and Li-ion-conducting compounds can reduce direct contact with reactive sulfides, while artificial anode interlayers can redistribute current and accommodate deformation. Coating benefits depend on conformal coverage and a controlled balance of transport and stability; overly thick or poorly conducting layers merely replace chemical instability with high impedance [5, 22]. Gradient structures and compliant interlayers are promising because they can address chemical and mechanical mismatch simultaneously.

4.4. Composite electrolyte engineering

Composite electrolytes combine phases with complementary functions. In polymer–ceramic systems, the polymer provides flexibility and processability, while the ceramic filler can suppress crystallization, reinforce the membrane, or create additional transport pathways [11, 12]. The outcome depends on filler chemistry, surface state, orientation, percolation, polymer–salt coordination, and the resistance of the polymer–ceramic interface.

Hybrid composites incorporating oxides, sulfides, polymers, or functional nanomaterials broaden the design space but also introduce additional internal interfaces. Achieving homogeneous dispersion, reproducible thickness, low interfacial resistance, and long-term phase compatibility remains difficult. Mechanism-guided design should therefore distinguish between passive reinforcement, interfacial transport enhancement, and true continuous inorganic conduction rather than attributing every improvement to generic synergy.

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4.5. Future perspectives for solid electrolyte design

No single electrolyte family or modification strategy simultaneously maximizes conductivity, electrochemical stability, mechanical robustness, processability, and low cost. Integrated optimization must connect composition, microstructure, interfaces, electrode architecture, and operating conditions [1, 2, 6, 19]. Comparisons should prioritize complete-cell metrics under realistic loading, thickness, pressure, temperature, and lithium-inventory conditions.

Computational screening and machine-learning methods can accelerate candidate selection, but their value depends on reliable structures, consistent property data, and experimental validation [33]. Operando and depth-resolved characterization are likewise essential for separating bulk, grain-boundary, and interfacial degradation [13, 14]. The most productive workflow is therefore iterative: computation identifies candidates, controlled synthesis tests them, multiscale characterization reveals failure mechanisms, and the resulting data refine subsequent design.

5. Outlook and conclusion

Solid-state batteries have moved beyond the search for fast ion conductors and are now being judged as complete electrochemical systems. The literature reviewed here makes clear that bulk conductivity, although essential, is not a sufficient predictor of cell performance. Transport through composite electrodes, reactions at buried interfaces, defect populations, lithium morphology, pressure, and manufacturing variability act together and often become more restrictive as cells approach practical dimensions. Recent research accordingly places greater emphasis on interface engineering, dendrite suppression, stack pressure, and practical cell design [1, 2, 14, 19].

The major electrolyte families should be viewed as different sets of compromises. Sulfides combine high conductivity with favorable deformability, but moisture sensitivity and electrode reactivity complicate processing and stability. Oxides are generally more robust, yet their rigidity, grain-boundary resistance, and demanding densification can produce poor contact and higher cost. Polymers are attractive for thin-film processing and mechanical compliance, but room-temperature transport remains limiting. Composite electrolytes may balance some weaknesses, although their additional internal interfaces make uniform fabrication more difficult. No class currently satisfies all practical requirements without careful control of composition, microstructure, and cell architecture.

Interfaces will remain the central link between materials development and durable cell operation. Protective coatings and artificial interlayers can limit parasitic reactions, but they must remain ionically conductive and mechanically intact during repeated volume changes. At the lithium-metal side, suppressing short circuits requires more than a stiff separator. Void formation during stripping, nonuniform plating, surface flaws, grain boundaries, and local stress all influence filament growth. Design rules must therefore connect interphase chemistry with contact mechanics and current distribution under realistic temperature and pressure, rather than treating chemical and mechanical degradation separately.

Future studies would benefit from a more consistent basis for comparison. Reports of conductivity, critical current density, or cycle life should include electrolyte thickness, cathode areal loading, lithium inventory, current density, temperature, stack pressure, active area, and cell-to-cell variation. Testing thin electrolytes and high-loading cathodes with limited lithium excess may give less striking initial results, but it reveals failure modes that matter at scale. Computation can narrow the search space if datasets are consistent and experimentally verified. Operando and depth-resolved characterization should then identify whether performance loss begins in the bulk, grain boundaries, reaction layers, or physical contact loss.

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Materials discovery and manufacturing development must proceed together. Processing routes need to address thin-membrane quality, moisture control, powder handling, lamination, and low-pressure operation; a material that performs well only as a thick pellet or under very high pressure is unlikely to retain the same advantage in a large-format cell. Commercial progress will therefore come from steady gains across coupled variables rather than from one record material. The most credible path combines stable chemistry, controlled microstructure, resilient interfaces, balanced electrode architecture, reproducible manufacturing, and transparent cell-level testing. Safety and cost should be assessed alongside electrochemical performance. Solid-state batteries remain promising, but success will depend on preserving material-level advantages in thin, high-loading, manufacturable cells.

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