

Phase Stability and Electrical Performance of Solid-State Battery Materials

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ABSTRACT

The escalating demand for safer, higher-energy-density energy storage solutions has positioned solid-state batteries (SSBs) as a promising successor to conventional lithium-ion batteries, addressing critical limitations such as flammability, limited energy density, and safety risks associated with liquid electrolytes. This review comprehensively examines the phase stability and electrical performance of key solid electrolyte materials, emphasizing thermodynamic principles governing interfacial reactions, ion transport mechanisms, and strategies to overcome solid-solid interface challenges in all-solid-state systems. Thermodynamic stability is evaluated through lithium chemical potential windows and grand potential phase diagrams, revealing that decomposition driving forces dictate compatibility with high-voltage cathodes and lithium-metal anodes. Oxide-based electrolytes (garnet LLZO, NASICON LATP) offer wide electrochemical windows (>5 V) and excellent chemical/thermal stability but suffer from brittleness and high interfacial resistance. Sulfide electrolytes (LGPS, argyrodites) deliver superionic conductivities (up to 25 mS/cm) and mechanical ductility for facile processing, yet face narrow stability windows, air/moisture sensitivity, and H_2S evolution risks. Emerging halide electrolytes and high-entropy materials provide balanced oxidative stability and improved low-temperature performance, while polymer and composite electrolytes enhance flexibility and wettability, albeit with lower room-temperature conductivity. Electrical performance hinges on ionic conduction pathways (Arrhenius behavior, BCC frameworks), grain boundary contributions, electronic insulation to prevent self-discharge, and charge-transfer kinetics at rigid interfaces. Interphase engineering via coatings (e.g., LiNbO_3 , Al_2O_3), doping, and artificial interlayers mitigates resistive or mixed-conducting interphases. Electro-chemo-mechanical stresses from volume changes (up to 18% in high-energy cells) are addressed through compliant binders and reduced stack pressures. Advanced operando characterization and machine learning accelerate material discovery and performance prediction. Scalable manufacturing roadmaps project significant cost reductions ($<\$100/\text{kWh}$ by 2035) and market growth, with pilot production and automotive integration targeted for 2027–2028. Despite persistent hurdles in interfacial stability, mechanical integrity, and low-temperature operation, integrated strategies combining material innovation, interface engineering, and computational design pave the way for commercially viable SSBs with superior

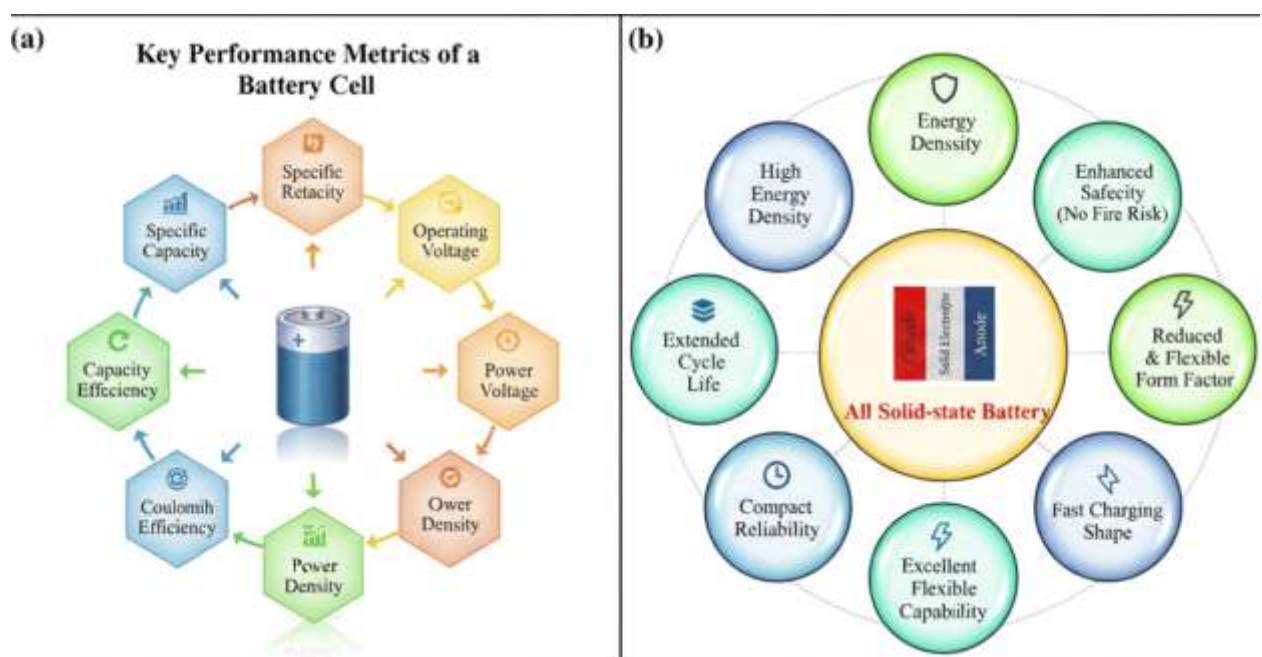
safety, energy density (>400 – 500 Wh/kg), and cycle life.

Keywords: Solid-State Batteries, Phase Stability, Solid Electrolytes, Ionic Conductivity, Interfacial Stability, Lithium-Metal Anode, Grand Potential Phase Diagram, Oxide Electrolytes, Sulfide Electrolytes, Halide Electrolytes, High-Entropy Materials, Electro-Chemo-Mechanical Coupling, Interphase Engineering, All-Solid-State Lithium Batteries

Introduction

The global transition toward sustainable energy systems has placed electrochemical energy storage at the center of modern technological development. Conventional lithium-ion batteries (LIBs), while successful in personal electronics and early-stage electric vehicles, are increasingly scrutinized for their inherent safety limitations and the plateauing of their energy density (Hamdan et al., 2024). The reliance on flammable organic liquid electrolytes creates significant risks of thermal runaway, particularly when utilized alongside high-capacity electrodes or operated under extreme conditions. Solid-state batteries (SSBs) have emerged as the definitive successor, promising a transformative leap in safety, specific energy, and operational lifespan (Alsaç et al., 2025). By replacing volatile liquid components with inorganic or polymer-based solid electrolytes, these systems mitigate combustion risks and potentially enable the integration of lithium-metal anodes and high-voltage cathodes (Shen et al., 2026). However, the realization of commercially viable all-solid-state batteries is predicated on solving profound challenges related to the thermodynamic phase stability of materials and the complex electrical performance at rigid solid-solid interfaces (Xiao et al., 2020).

Figure 2. (a) Key Performance Metrics of a Battery Cell and (b) Strategic Advantages of Transitioning to All-Solid-State Configurations.



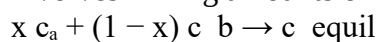
Thermodynamic Foundations of Phase Stability

The thermodynamic stability of a solid electrolyte in contact with an electrode is the primary factor determining the feasibility of a battery chemistry (Tan et al., 2025). Unlike liquid electrolytes, which can conform to surface irregularities and often form a self-limiting interphase, solid-state systems are characterized by rigid boundaries where chemical reactivity can lead to high-impedance reaction layers or mechanical delamination (Nie et al., 2023). Research indicates that the bulk thermodynamic driving force for reactions between the electrolyte and the cathode is a robust proxy for interfacial stability, often exerting a more significant influence than interfacial energy itself (Zhu et al., 2015).

Grand Potential and Lithium Chemical Potential Windows

The evaluation of interfacial stability is traditionally conducted through the lens of the lithium chemical potential stability window. This window defines the range of potentials over which the solid electrolyte remains electrochemically inert (Chen et al., 2022). If the chemical potential of the cathode exceeds the oxidative limit of the electrolyte, or if the anode potential falls below the simple potential window analysis is often insufficient to capture the complexity of multi-component reactions at the interface (Wei et al., 2025).

To address this, computational models utilize the grand potential phase diagram, which treats the system as being open to lithium. This is represented by the function $\Phi[c, \mu_{\text{Li}}]$, where the ground state structure or phase equilibrium is determined at a given composition and lithium potential (Al Awad, 2025). The reaction energy is calculated by finding the products that form with the largest driving force when combining the electrolyte and electrode phases (Carrasco, 2024). The general reaction involves mixing amounts of each phase:



Where x is a mixing parameter and c_{equil} represents the low-energy equilibrium determined from the phase diagram. The resulting change in grand potential determines the interfacial stability as a function of the average cathode voltage (Cao et al., 2024). The most successful interfaces are those where the decomposition products are ionically conductive but electronically insulating, effectively passivating the surface and preventing further electrolyte consumption (Guo et al., 2022).

Thermodynamic Driving Forces in Cathode Compatibility

The interaction between the electrolyte and high-voltage cathodes is particularly aggressive. Many sulfide-based electrolytes possess a narrow electrochemical window, making them susceptible to oxidation when paired with lithium-rich or high-nickel oxides (Man et al., 2025). The thermodynamic driving force for these reactions can be predicted by the difference in chemical potential between the pristine phases and the equilibrium decomposition products. For example, sulfide electrolytes often decompose into elemental sulfur, phosphorus sulfides, and metal sulfides, some of which may be ionically resistive and hinder charge transfer (Lu et al., 2022). Conversely, oxide-based electrolytes generally exhibit much wider thermodynamic stability windows, which explains their compatibility with high-voltage cathodes without the immediate formation of resistive interphases (Eng, 2021).

The Solid Electrolyte Portfolio: A Mechanistic Review

Solid-state electrolytes are broadly classified based on their chemical composition and the mechanisms by which they facilitate ion transport. The three primary classes are inorganic solid electrolytes (including oxides, sulfides, and halides), organic polymer electrolytes, and composite systems (Yang & Wu, 2020).

Oxide-Based Solid Electrolytes

Oxide electrolytes are characterized by their exceptional chemical and thermal stability. Leading materials include garnet-type $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO), NASICON-type $\text{Li}_{1+x}\text{Al}_x\text{Ti}_{2-x}(\text{PO}_4)_3$ (LATP), and perovskite-type $\text{Li}_{3x}\text{La}_{2/3-x}\text{TiO}_3$ (LLTO). These materials are generally non-flammable and exhibit very wide electrochemical stability windows, often exceeding 5.0 V vs. Li/Li^+ (Primo, 2025).

Table 1. Characteristics and Performance Metrics of Major Solid-State Electrolyte Categories.

Electrolyte Category	Representative Materials	Ionic Conductivity (mS/cm)	Advantages	Limitations
Garnet Oxide	LLZO, LLZTO	0.1 - 1.0	Wide window, stable vs. Li	Brittle, high sintering temp
NASICON Oxide	LATP, LAGP	0.1 - 1.0	Air stable, high conductivity	Unstable vs. Li metal
Sulfide	$\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS)	12 - 25	Superionic conductivity, soft	Moisture sensitive, narrow window
Argyrodite	$\text{Li}_6\text{PS}_5\text{Cl}$	1.0 - 5.0	Facile processing, ductile	H_2S evolution risk
Halide	Li_3InCl_6 , Li_3YCl_6	0.1 - 2.0	Stable vs. high-voltage cathodes	Moisture sensitive
Polymer	PEO + LiTFSI	0.0001 to 0.01 (at 60C)	Flexible, scalable	Low RT conductivity, soft

The primary challenge with oxide electrolytes is their mechanical rigidity and the high temperature required for processing. Sintering temperatures typically range from 1000 to 1200 degrees Celsius, which can lead to deleterious side reactions if the electrolyte is co-sintered with the cathode (Balaish et al., 2025). Furthermore, oxides are brittle, leading to poor interfacial wetting and high area-specific resistance (Zakaria et al., 2020).

Sulfide-Based Solid Electrolytes

Sulfide electrolytes are often considered the most promising candidates for high-power applications due to their superionic conductivity. Materials such as $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS) have demonstrated conductivities as high as 25 mS/cm,

surpassing even commercial liquid electrolytes (Kato et al., 2020). The high conductivity is attributed to the larger, more polarizable sulfur ions, which create a more open lattice for lithium migration (Karabelli et al., 2021).

Mechanically, sulfides are much softer and more ductile than oxides, allowing them to be processed via cold-pressing at room temperature (Oh et al., 2025). This ductility facilitates excellent particle-to-particle contact within composite electrodes. However, sulfides suffer from poor chemical stability in air, reacting with moisture to release toxic H₂S gas (Kim et al., 2021). Their electrochemical window is also relatively narrow, often requiring protective coatings to prevent continuous decomposition (Nikodimos et al., 2022).

Halide and High-Entropy Electrolytes

Halide electrolytes, such as Li₃InCl₆ and Li₃YCl₆, have emerged as a high-performance middle ground. They offer oxidative stability superior to sulfides, allowing for direct integration with nickel-rich cathodes like NMC811 without coatings (Tuo et al., 2023). Recent breakthroughs in mixed-anion halides, such as Li_{1.6}AlCl_{3.4}SO_{0.6}, have pushed ionic conductivity to 0.18 mS/cm at 25 degrees Celsius, offering a cost-effective alternative to rare-earth halides (Jun, 2024).

High-entropy materials (HEMs) represent a new paradigm in electrolyte design. By incorporating multiple elements into a single lattice, configurational entropy is used to suppress phase separation and lattice distortion (Chen et al., 2025). High-entropy garnet-type oxides have demonstrated lithium-ion conductivity of 0.62 mS/cm at temperatures as low as -60 degrees Celsius, enabling all-solid-state cells with 91% capacity retention after 100 cycles in extreme environments (Stockham et al., 2022).

Solid Polymer and Composite Electrolytes

Organic solid polymer electrolytes (OSPEs) are typically composed of high molecular weight polymers like poly (ethylene oxide) (PEO) and lithium salts. They offer excellent flexibility and processability, but their ionic conductivity at room temperature is insufficient for most applications (An et al., 2022).

Composite solid electrolytes (CSEs) combine the advantages of inorganic fillers and polymer matrices. The integration of oxide or sulfide particles into a polymer framework can enhance mechanical strength and improve the transference number via space-charge layer mechanisms (Liu et al., 2023). CSEs are particularly effective at mitigating the high interfacial resistance found in purely inorganic systems by providing a more compliant contact with the electrodes (Khan et al., 2024).

Electrical Performance and Ion Transport Dynamics

The electrical performance of solid-state materials is defined by the efficient transport of ions and the effective blockage of electrons. The interplay between bulk conductivity, grain boundary resistance, and electronic leakage determines the power density and self-discharge characteristics of the cell (Xiong et al., 2025).

Mechanisms of Ionic Conduction

In crystalline solid electrolytes, ion migration is a thermally activated process governed by the ability of charge carriers to move between stable ground-state sites

and intermediate metastable sites within an anionic framework (Mulryan & Islam, 2019). This behavior is commonly described by a modified Arrhenius-type relationship:

$$\sigma = A T^m \exp\left(-\frac{E_a}{k_B T}\right)$$

where σ is the ionic conductivity, A is the pre-exponential factor, T is the absolute temperature, m is a temperature-dependent exponent related to lattice dynamics, E_a is the activation energy for ion migration, and k_B is the Boltzmann constant. The underlying crystal structure strongly influences ion transport. In particular, body-centered cubic (BCC) anionic frameworks are often favorable for fast ion conduction because they enable direct hopping pathways between tetrahedral sites, thereby avoiding energetically unfavorable tetrahedral–octahedral–tetrahedral migration pathways (Sau et al., 2024). Such structural motifs reduce migration barriers and enhance ionic conductivity.

Grain Boundary and Electronic Resistance

In polycrystalline materials, the total ionic resistance is a combination of bulk and grain boundary contributions. Grain boundaries are often regions of structural disorder and local strain, leading to higher activation energies and lower conductivity compared to the bulk (Quirk et al., 2024). A critical requirement for any solid electrolyte is high electronic resistance. Low electronic resistance leads to self-discharge and the internal reduction of lithium ions, which can manifest as dendrite growth within the electrolyte (Vahidi et al., 2021).

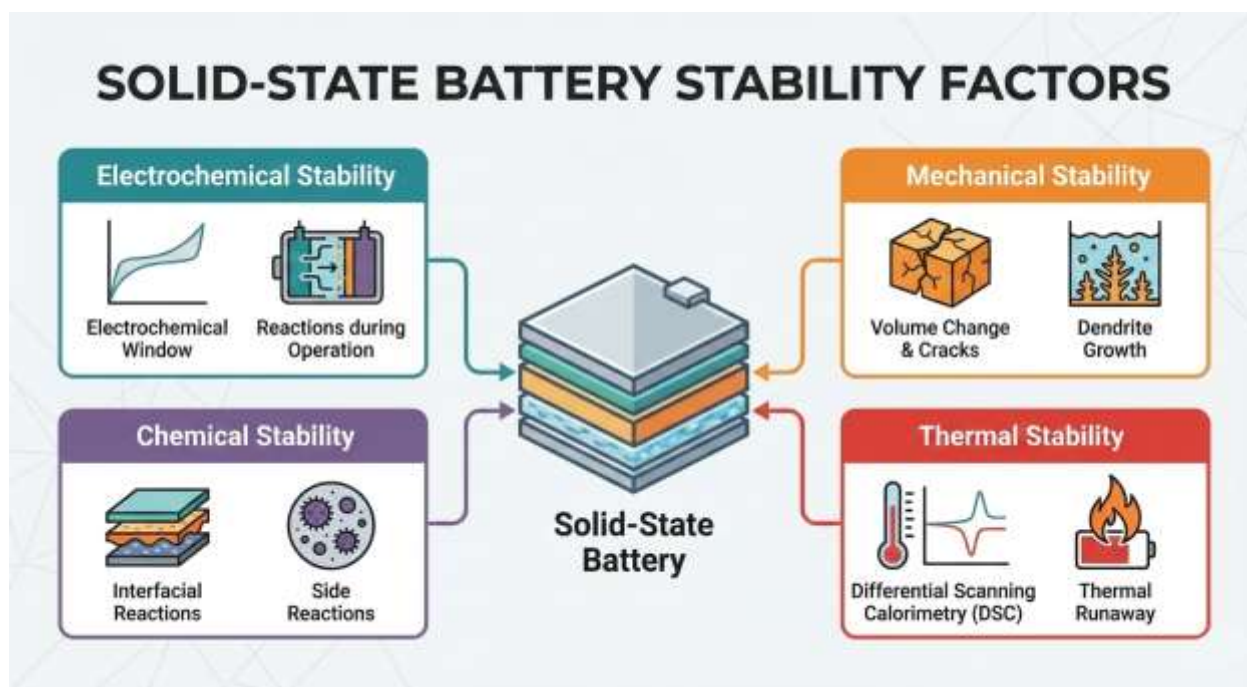
Charge Transfer Kinetics and Overpotentials

The driving force for redox reactions in a battery is the difference in electrochemical potential between phases. In solid-state systems, the magnitude and spatial distribution of the potential drop across an interface fundamentally influence the charge-transfer kinetics. The exchange current density at the solid-solid interface is often lower than in liquid electrolytes due to limited contact area and resistive interphases (Weiss et al., 2020). Researchers have employed operando techniques to monitor shifts in kinetic energy during charging, providing a direct measure of the electrostatic potential drop (Krauskopf et al., 2020).

Interfacial Stability and Interphase Engineering

The buried interfaces in solid-state batteries are the primary sites of performance degradation. Stability at these boundaries is categorized into chemical stability and electrochemical stability (Banerjee et al., 2020).

Figure 3. Critical Stability Pillars in Solid-State Batteries: Electrochemical, Chemical, Mechanical, and Thermal Considerations.



Solid-Electrolyte Interphase (SEI) and Cathode-Electrolyte Interphase (CEI)

When a solid electrolyte is in contact with an electrode outside its stability window, it decomposes to form an interphase layer. These are typically classified into three types: stable passivating interphases (Type I/III), and Mixed Conducting Interphases (Type II) (Xiao et al., 2020). In sulfide systems, reactions with lithium metal often form mixed conducting interphases containing Li_3P and Li_2S , which can lead to continuous interphase growth and capacity fade (Liang et al., 2022).

Surface Modification and Coating Strategies

To stabilize these interfaces, several engineering strategies have been developed:

Artificial Interlayers: Depositing thin, stable layers (e.g., LiNbO_3 , Al_2O_3) onto the cathode surface to prevent the oxidation of sulfide electrolytes (Alsaç et al., 2025).

Doping: Introducing elements like Sc or Ca into the electrolyte structure to tune the redox potential and enhance stability against lithium metal (Chen et al., 2025).

Wettability Enhancements: For lithium-metal anodes, interfacial layers of Ag or Zn can be used to form a transient alloy, ensuring more uniform lithium deposition (Lee et al., 2020).

Electro-Chemo-Mechanical Integrity and Volume Regulation

In solid-state systems, volume changes generate significant internal stresses that can lead to fracture and contact loss. High-nickel cathodes like NMC811 typically expand and contract by about 6% (Oh et al., 2025). In high-energy configurations targeting 600 Wh/kg, the overall cell-level volume change can reach 18%, causing localized stresses in the gigapascal (GPa) range (Nikodimos et al., 2022).

These changes induce several failure modes, including interfacial delamination and active material fracture (Nie et al., 2023). To maintain contact, laboratory cells often

use high stack pressures, but commercial vehicular applications require operation below 1 MPa (Oh et al., 2025). Strategies to mitigate these issues include the use of elastomeric binders (e.g., poly (carbonate-b-ether-b-carbonate)) in composite cathodes to buffer volume changes (Yang & Wu, 2020).

Thermal Stability and Runaway Dynamics

The non-flammable nature of solid electrolytes is a primary safety advantage. Oxide electrolytes generally remain stable beyond 1000 degrees Celsius, while sulfide-based composites can reach thermal decomposition temperatures above 500 degrees Celsius (Karabelli et al., 2021). Polymer electrolytes based on PEO are more limited, typically beginning to decompose between 160 and 400 degrees Celsius depending on the lithium salt used (Man et al., 2025). The elimination of the liquid shuttle effect in SSBs provides a rigid physical barrier that prevents the immediate cascading of thermal events during a short circuit (Xiao et al., 2020).

Table 2. Typical Thermal Decomposition Temperatures and Failure Mechanisms of Battery Materials.

Material Class	Typical Decomposition Temp (C)	High-Temperature Failure Mechanism
Pure PEO	400	Structural collapse, melting
PEO/LiBF ₄	160	Salt-driven decomposition
PEO/LiTFSI	300	Gradual degradation
Sulfide/Polyimide	> 500	Composite structural breakdown
Halide (LOHC)	320	Melting/increased reactivity in liquid phase
LLZTO/PVDF	310	Polymer phase failure

Advanced Characterization and Diagnostic Methodologies

Synchrotron radiation provides the high photon flux necessary for deep probing of solid cells. Operando X-ray diffraction mapping has unveiled heterogeneous lithiation and delithiation processes within individual cathode particles (Primo, 2025). Additionally, the PEEK in-situ Press (PIP) cell enables 3D visualization of internal structures during cycling with a resolution of 750 nm, revealing the propagation of defects and stress-induced cracking in solid electrolyte pellets (Young et al., 2025).

Figure 1. Multiscale Modeling, Characterization, and Diagnostic Framework for Solid-State Battery Development.

Machine Learning and Computational Discovery

Artificial intelligence (AI) has become an indispensable tool in SSB research, moving from a post-hoc analysis tool to a real-time copilot. Generative models like MatterGPT allow for the inverse design of materials, targeting specific properties such as formation energy and band gaps (Eng, 2021). ML technology also accurately predicts key performance indicators; hybrid CNN-LSTM models have achieved less than 1% error in state-of-charge (SOC) estimation under dynamic loads, while graph convolutional networks forecast remaining useful life (RUL) with an error rate as low

as 0.14% (Wang & Yu, 2025).

Scalable Manufacturing and Industrial Roadmaps

The transition of solid-state technology to high-volume manufacturing (HVM) is a critical challenge. The year 2025 has been identified as a pivotal milestone for industrialization, particularly in the Asia-Pacific region. Companies like Samsung and Nissan have announced plans for mass production and automotive installation by 2027–2028, with cost targets as low as 65 dollars per kilowatt-hour (Beyer, 2025).

Table 3. Global Solid-State Battery Market Projections and Key Attributes (2025–2035).

Market Attribute	2025 Projection	2035 Projection
Market Size (USD)	~1.67 Billion	~48.2 Billion
Dominant Segment	Bulk SSB (~86%)	Bulk SSB (EVs)
Production Cost	400 - 800 per kWh	< 100 per kWh (Target)

Conclusion

Solid-state batteries represent a paradigm shift in electrochemical energy storage, offering inherent safety advantages through non-flammable electrolytes, compatibility with high-capacity lithium-metal anodes, and potential for dramatically higher energy densities compared to traditional lithium-ion systems. This review highlights that thermodynamic phase stability governed by lithium chemical potential windows and decomposition driving forces remains the foundational criterion for material selection and interfacial design, while electrical performance is ultimately limited by ion transport efficiency, grain boundary resistance, electronic leakage, and charge-transfer overpotentials at solid-solid contacts. Significant progress across electrolyte classes demonstrates trade-offs: oxides provide unmatched stability but demand sophisticated processing and interface mitigation; sulfides excel in conductivity and processability yet require protective strategies against oxidation and environmental degradation; halides and high-entropy compositions emerge as compelling compromises with enhanced oxidative tolerance and low-temperature capabilities; and composites/polymers offer mechanical compliance at the expense of bulk conductivity. Effective solutions to persistent challenges such as high interfacial impedance, dendrite formation, volume-induced stresses, and manufacturing scalability are advancing through targeted interphase engineering, doping, coatings, compliant architectures, and AI-driven discovery. Recent milestones in operando diagnostics, high-entropy material design, and industrial roadmaps underscore accelerating progress toward practical deployment, with projections indicating market expansion and cost-competitive production in the coming decade. Ultimately, realizing the full promise of solid-state batteries will require holistic integration of thermodynamic understanding, kinetic optimization, mechanical resilience, and scalable fabrication. Continued interdisciplinary efforts in materials science, computational modeling, and engineering will be essential to overcome remaining barriers, enabling safer, longer-lasting, and higher-performance energy storage systems critical for sustainable electrification, grid-scale renewables, and next-

generation mobility.

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