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Toward a Unified Mechanistic Understanding of Polymer Electrolytes for Advanced Solid-State Batteries

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ABSTRACT

Polymer electrolytes (PEs) are widely regarded as a promising platform for solid-state batteries (SSBs), offering the potential to simultaneously achieve high energy density with improved safety. However, in current literature, PEs spanning liquid-percolated gels, liquid-assisted quasi-solids, and truly polymer-governed solids are often indiscriminately grouped as solid polymer electrolytes (SPEs), obscuring their distinct ion transport mechanisms, interfacial behaviors, and practical performance constraints, and leading to misleading performance comparisons and unrealistic expectations regarding solid-state operation. Herein, we establish a mechanistic framework that categorizes PEs into gel polymer electrolytes (GPEs), quasi-solid polymer electrolytes (QSPEs), and all-solid polymer electrolytes (ASPEs) based on their dominant ion-solvation environment and transport pathways. By systematically analyzing the ion-transport mechanisms, interfacial behaviors, and performance-limiting features associated with each PE class, we clarify their defining characteristics and mechanism-imposed limitations. Accordingly, we outline *category-specific* research priorities and highlight the necessity of mechanism-driven materials design, transparent definitions and reporting, and application-relevant benchmarking. This unified Perspective lays a foundation for consistent interpretation, meaningful comparison across PE systems, and more rational materials design toward the advancement of PE-enabled SSBs.

Jing Chen and Han Chen contributed equally to this work.

1 | Introduction

The accelerating demand for high-performance energy-storage technologies has intensified the pursuit of battery technologies capable of simultaneously delivering higher energy density and enhanced operational safety [1, 2]. While intercalation-based lithium-ion batteries (LIBs) have achieved remarkable commercial success, their energy density is approaching their practical limits [3, 4]. Further breakthroughs require integrating lithium metal anodes and high-voltage cathodes [5]. However, these high-energy electrode chemistries pose severe challenges to conventional liquid electrolytes. Strong chemical reactivity and electrochemical instability at electrode–electrolyte interfaces give rise to parasitic reactions, safety risks, and rapid performance degradation even under non-aggressive operating conditions [6–8]. These limitations have motivated intensive interest in solid-state electrolytes (SSEs), which can mitigate liquid electrolyte-driven interfacial instability while enabling safer operation and compatibility with lithium metal anodes and high-voltage cathodes [9–13].

Among various SSEs, polymer electrolytes (PEs) have attracted considerable attention owing to their mechanical flexibility, processability, and chemical designability (Figure 1) [14–17]. Notably, Bolloré Group has demonstrated practical large-scale deployment of SPE-based solid-state lithium metal batteries (SSLMBs), with cumulative deployed capacities exceeding 1 GWh since 2010, enabling applications in both electric vehicles and grid-scale energy-storage systems [18]. However, classical poly(ethylene oxide) (PEO)-based PEs depend on segmental-motion-governed ion transport, which limits room-temperature (RT) ionic conductivity and often requires elevated-temperature operation [12, 19]. This constraint has hindered the practical realization of RT all-solid-state batteries (ASSBs) based on PEs. To overcome this limitation, extensive research has focused on enhancing RT ionic conductivity through the incorporation of plasticizers or other functional additives into polymer matrices [16, 20–23]. These plasticizers broadly include liquid or liquid-like components, such as small-molecule solvents [24, 25], ionic liquids or molten salts [22, 26, 27], and even liquid electrolytes [28, 29], as well as solid organic plastic crystals [30–32]. While such strategies can enhance RT ionic conductivity by 2–3 orders of magnitude in otherwise poorly conducting polymer systems, they can give rise to distinct ion-transport mechanisms depending on the nature of additives [33–37]. *Specifically, when plasticizers generate percolating liquid phases, Li^+ transport becomes dominated by the diffusion of solvated species through the liquid phase, corresponding to liquid-dominated transport* [24–26, 28, 29]. *When such species remain non-percolating, Li^+ migration proceeds through dynamically mobile liquid-like species that coordinate Li^+, constituting a liquid-assisted pathway* [31, 38–41]. B*y contrast, when solid plasticizers primarily enhance polymer segmental dynamics without introducing liquid-mediated vehicular pathways, long-range Li^+ transport remains predominantly polymer-governed* [22, 30, 36, 37, 42].

Despite these distinct transport regimes, PEs containing substantial liquid or liquid-like components are frequently grouped under the label of *solid polymer electrolytes (SPEs) based solely on macroscopic appearance* [17, 20, 23, 43–48], thereby overlooking *fundamental differences in ion-transport behavior and leading to unrealistic performance comparisons that overstate progress in truly polymer-governed systems*. In essence, PEs should be viewed as a compositional and mechanistic continuum extending from liquid-dominated gels, liquid-assisted quasi-solids, to genuinely polymer-governed solids. *Because these distinct transport mechanisms dictate different safety characteristics, interfacial chemistries, and cell-level constraints, meaningful comparisons require explicit identification of the dominant conduction mechanism and the role of liquid or liquid-like components.*

In this Perspective, we establish a unified, mechanism-driven classification framework that categorizes PEs into gel polymer electrolytes (GPEs), quasi-solid polymer electrolytes (QSPEs), and all-solid polymer electrolytes (ASPEs) based on *their dominant ion transport pathways* (Figure 1). *Specifically, GPEs exhibit liquid-dominated vehicular transport enabled by percolating liquid phases; QSPEs display liquid-assisted transport through essential but non-percolating liquid-like species; and ASPEs rely on polymer-governed Li^+ solvation and migration without dependence on liquid-mediated vehicular pathways.* We further analyze the defining features and key limitations of each category, identify persistent misconceptions in the literature, and outline research priorities essential for advancing PEs toward genuinely competitive solid-state battery technologies. By promoting clearer terminology, standardized reporting practices, mechanism-centered materials design, and application-relevant benchmarking protocols, this Perspective aims to support more coherent progress and more meaningful cross-study comparisons within the PE community. More broadly, the mechanistic principles and classification criteria discussed herein are applicable to PEs developed for other metal batteries, where analogous ambiguities in ion-transport mechanisms and solvent participation frequently arise [49–56].

2 | Classifications, Mechanisms, and Persistent Challenges of PEs

Accurate classification of PEs is essential for understanding their ion-transport behavior, diagnosing failure mechanisms, benchmarking electrochemical performance, and guiding the rational design of next-generation SSB materials. Such a framework requires identifying the dominant ion-transport pathway and the role of liquid or liquid-like species, thereby linking material composition to transport mechanism, electrochemical behavior, and practical limitations. On this basis, PEs can be differentiated into GPEs, QSPEs, and ASPEs, each of which exhibits distinct transport characteristics, interfacial behaviors, and associated challenges.

2.1 | Gel Polymer Electrolytes (GPEs)

GPEs represent polymer-liquid hybrid systems in which a three-dimensional polymer network immobilizes a substantial fraction of liquid components, thereby forming a liquid-percolated system. In such systems, the polymer matrix primarily provides mechanical integrity and dimensional stability, whereas long-range ion transport is dominated by the percolating liquid phase [57–60]. Lithium ions (Li^+) predominantly reside in liquid solvent-coordinated states and migrate through interconnected liquid domains via diffusion of solvated species, closely

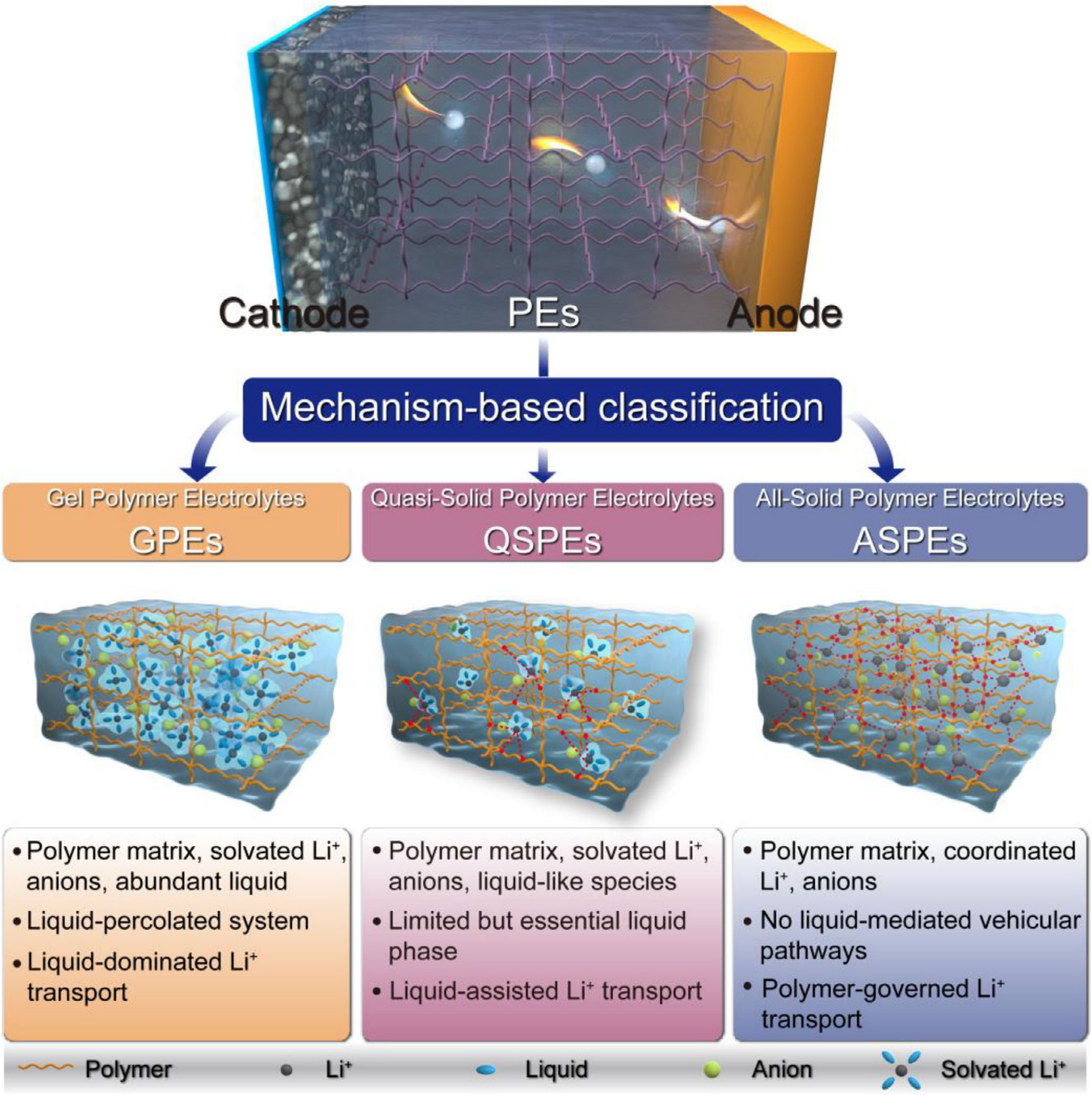

FIGURE 1 | Mechanistic classification of polymer electrolytes (PEs). PEs are categorized into gel polymer electrolytes (GPEs), quasi-solid polymer electrolytes (QSPEs), and all-solid polymer electrolytes (ASPEs) according to their dominant Li^+ solvation environments and transport pathways. In GPEs, Li^+ transport is governed by liquid-dominated vehicular diffusion through continuous, percolating liquid domains confined within a polymer network. In QSPEs, Li^+ solvation and long-range transport rely on a limited yet essential fraction of dynamically mobile liquid-like species, resulting in liquid-assisted diffusion within a macroscopically solid matrix. In ASPEs, both Li^+ solvation and long-range transport are governed predominantly by polymer coordination and segmental dynamics, without participation of liquid-mediated vehicular pathways.

resembling the ion transport behavior in conventional liquid electrolytes (Figure 2a). Owing to a liquid-dominated vehicular transport mechanism, GPEs typically exhibit high RT ionic conductivity of 10^{-3}–10^{-2} S cm^{-1}, substantially exceeding those of other PE classes [34, 61, 62]. Compared with conventional liquid electrolytes, the polymer network provides mechanical integrity and confinement of the liquid phase, thereby reducing leakage and partially mitigating volatility and flammability [59, 63, 64]. Importantly, although ion transport in GPEs is largely governed by the liquid phase, the structure of the polymer matrix can still influence transport indirectly by restricting anion mobility, regulating solvation structure, shaping interfacial ion flux, and stabilizing electrode–electrolyte interfaces [65, 66]. Depending on their chemical functionality, polymer matrices may either interact actively with solvated species or serve primarily as mechanically supporting frameworks. Moreover, the combination of a flexible polymer matrix and abundant liquid content affords excellent electrode interfacial wettability and

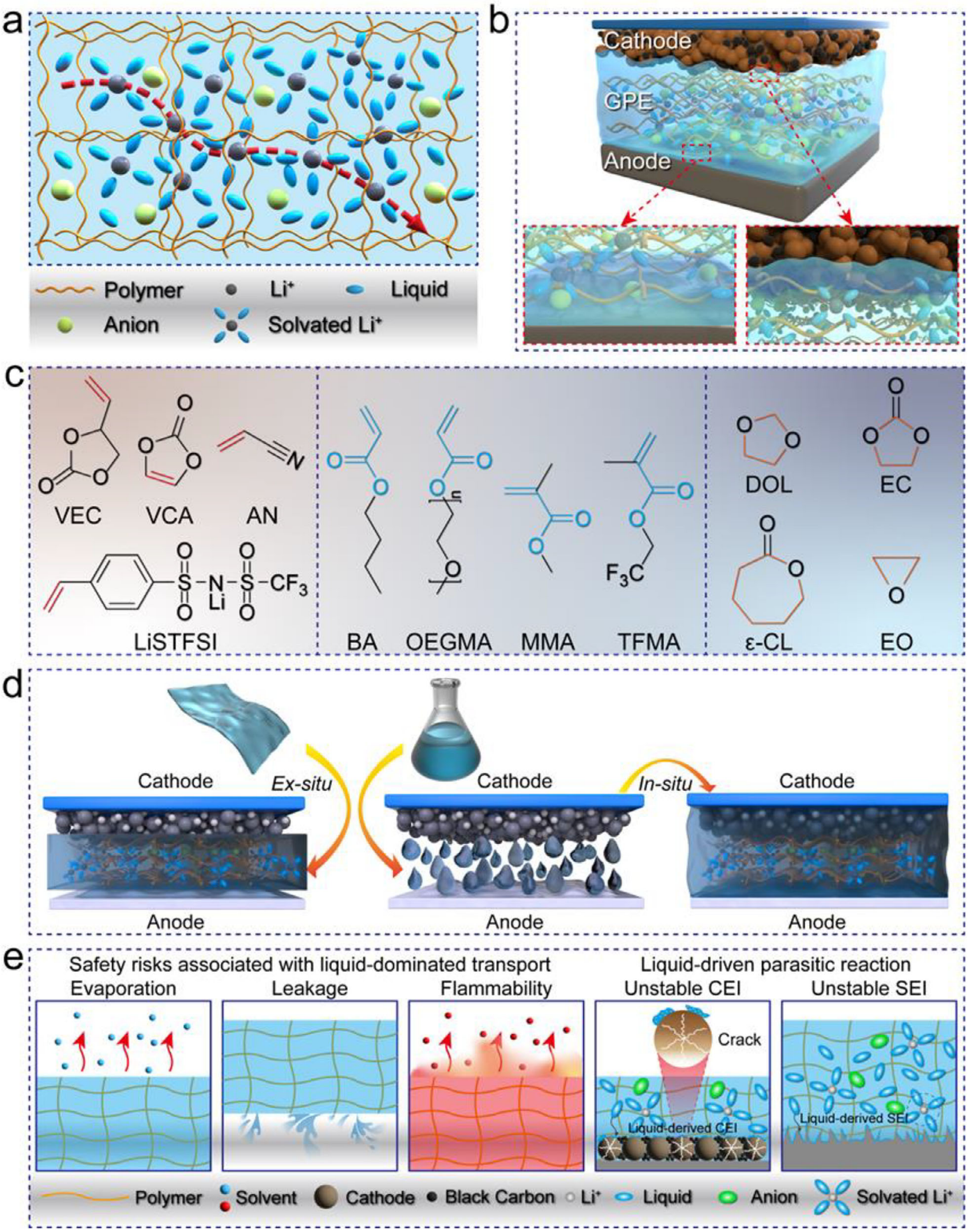

FIGURE 2 | Liquid-dominated ion transport mechanism and associated challenges in GPEs. (a) Ion transport in GPEs, where Li^+ migration is dominated by diffusion through percolated liquid domains confined by a mechanically supporting polymer network; (b) Liquid-mediated ion transport across bulk electrolyte and electrode interfaces in a full-cell configuration, illustrating enhanced interfacial wetting and ionic accessibility enabled by liquid-continuous pathways; (c) Representative polymerizable monomers employed in GPE systems, including unsaturated monomers, acrylate derivatives, and cyclic monomers; (d) Comparison between ex situ fabricated and in situ polymerized GPEs, highlighting improved electrolyte infiltration and electrode–electrolyte contact enabled by in situ polymerization; (e) Mechanism-associated constraints in GPEs arising from liquid-dominated transport, including safety concerns and liquid-driven interfacial parasitic reactivity reactions that lead to unstable CEI and SEI.

intimate interfacial contact, facilitating uniform Li deposition and effective electrolyte infiltration into high-loading porous cathodes [59, 61, 63]. These features collectively reduce interfacial resistance and enable electrochemical performance comparable to that of conventional liquid electrolyte systems (Figure 2b). From a physicochemical perspective, gelation in many GPEs can be viewed as thermodynamically driven phase separation (e.g., induced by temperature or solvent composition), leading to heterogeneous structures in which complete macroscopic phase separation is kinetically arrested. In this context, the sol–gel transition becomes a relevant descriptor of gel formation, independent of the absolute amount of immobilized liquid [47, 64, 67].

Representative GPEs are commonly synthesized through two main polymerization strategies. The first involves free-radical polymerization (FRP) of unsaturated vinyl monomers and polymerizable lithium salts or ionic liquids [68–73] including vinyl ethylene carbonate (VEC), vinylene carbonate (VCA), acrylonitrile (AN), and lithium (4-styrenesulfonyl)(trifluoromethanesulfonyl)imide (LiSTFSI), as well as a wide range of acrylate derivatives [74–77] such as butyl acrylate (BA), oligo(ethylene glycol) methyl ether methacrylate (OEGMA), methyl methacrylate (MMA), and trifluoroethyl methacrylate (TFMA). The second strategy relies on ring-opening polymerization (ROP) of cyclic monomers [78–80], including 1,3-dioxolane (DOL), ε-caprolactone (ε-CL), ethylene carbonate (EC), and ethylene oxide (EO), to form polymer networks that host liquid electrolytes (Figure 2c). GPEs can be obtained through either ex situ or in situ polymerization [29, 59, 63, 79, 81–83]. In ex situ approaches, a preformed polymer matrix absorbs liquid electrolyte, enabling precise control over polymer architecture and facilitating systematic material screening. However, such methods often suffer from limited interfacial conformity and slower electrolyte infiltration into porous electrodes. In contrast, in situ polymerization, where liquid monomers and salts are polymerized directly within the assembled cell, enables self-adaptive gel formation directly within the electrode structure, improving electrode–electrolyte contact and reducing interfacial impedance (Figure 2d). Despite these advantages, in situ polymerization routes are often constrained by cell-assembly conditions, reaction kinetics, and formulation complexity. *Incomplete monomer conversion and residual reactive species are frequently observed, which can compromise electrochemical stability, particularly at the Li metal interface*. These challenges underscore the importance of rigorously reporting monomer identity, molecular weight, crosslinker content, and polymerization conditions to ensure reproducibility and enable meaningful comparison across studies.

GPEs offer high RT ionic conductivity owing to their liquid-dominated ion-transport mechanism, making bulk ion transport rarely the primary performance-limiting factor. Despite being widely grouped under the SSEs category in the literature [17, 21, 34, 38, 46, 84], GPEs d*o not eliminate risks associated with liquid phases, including electrolyte evaporation or leakage, flammability, and liquid-driven parasitic reactions leading to unstable cathode-electrolyte interphase (CEI) and solid electrolyte interphase (SEI)* (Figure 2e) [60]. A persistent issue in current research is the tendency to overemphasize high ionic conductivity, while *far more critical challenges, such as safety, interfacial stability, and long-term mechanical reliability, are often underappreciated*. In many reported systems, apparent performance improvements primarily originate from the presence of liquid components rather than polymer-governed solid-state ion-transport behavior. In this context, GPEs are more appropriately considered as *mechanically solid in appearance but ionically liquid-dominated* systems in research claims, underscoring the importance of distinguishing them from *truly polymer-governed* solid electrolytes in performance and safety evaluation. Although GPEs strike a practical compromise between ion conductivity, interfacial contact, and manufacturability, *their liquid-dominated ion-transport mechanism renders electrochemical performance dependent on liquid-associated interfacial reactivity and safety constraints, thereby distinguishing them from genuinely polymer-governed solid-state systems*.

2.2 | Quasi-Solid Polymer Electrolytes (QSPEs)

QSPEs encompass systems in which a limited yet essential liquid-like species is constrained in the polymer matrix, despite exhibiting a macroscopically solid or self-supporting appearance. In typical QSPE systems, polar solvents or liquid plasticizers provide the primary Li^+ solvation environment, enabling ion transport predominantly through liquid-assisted diffusion of solvated species within polymer-confined domains (Figure 3a) [34, 85]. Crucially, unlike GPEs, these liquid-like species do not form a continuous, percolating phase, and therefore cannot support long-range liquid-phase diffusion, thereby differentiating QSPEs from GPEs. Representative QSPE systems include poly(vinylidene fluoride) (PVDF)- and cellulose-based electrolytes containing residual polar solvents or liquid plasticizers, where the polymer backbones exhibit weak Li^+ solvation capability and lack sufficient coordinating sites. Consequently, Li^+ ions are predominantly solvated by the residual solvent, most notably N,N-dimethylformamide (DMF), which dominates Li^+ coordination and forms solvated liquid-like species such as $[Li(DMF)_x]^+$ that mediate long-range ion transport (Figure 3b) [39, 40, 86–95]. The apparent ionic conductivity in these systems is therefore governed primarily by liquid-assisted diffusion through dynamically mobile liquid-like species, rather than by polymer-governed transport pathways.

Beyond conventional liquid-assisted QSPEs, a distinct subclass incorporates plastic-crystalline or eutectic-forming additives into otherwise ionically inert dry polymer matrices to create highly polar, dynamically disordered solvation environments that actively participate in Li^+ solvation and transport [12, 14, 31, 32, 43, 96, 97]. At s*ufficiently high additive and/or salt contents, strong coordination between these additives and lithium salts (LiTFSI) lowers the melting point and induces eutectic melting, giving rise to localized liquid-like regions that constitute plastic-crystal-derived eutectic or deep eutectic solvent (DES)-like solvation environments* (Figure 3c). *These confined solvation domains directly coordinate Li^+ and sustain liquid-assisted ion transport*, while remaining spatially confined within the polymer matrix and not establishing a percolating liquid phase. (Figure 3d) [41, 98, 99]. Representative plastic-crystalline or eutectic-forming additives used in QSPEs include succinonitrile (SN), *N*-methylurea (NMU), and *N*-methylacetamide (NMA), typically combined with LiTFSI to create highly polar, ion-conductive solvation

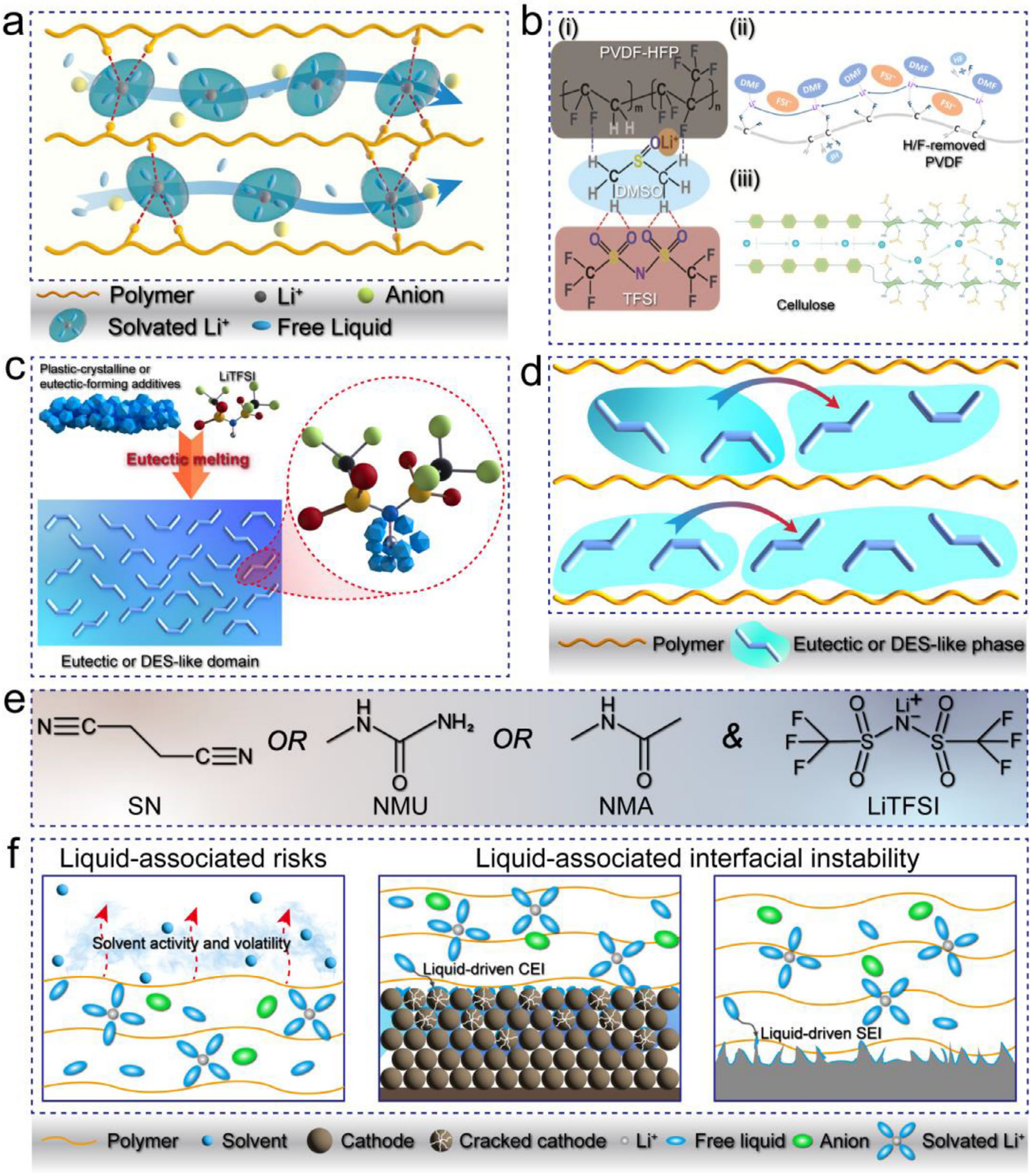

FIGURE 3 | Liquid-assisted ion transport mechanism and associated challenges in QSPEs. (a) Ion-transport mechanism in QSPEs, where long-range Li^+ migration is governed by liquid-assisted diffusion of mobile liquid-like species within polymer-confined domains; (b) Representative Li^+ coordination environment in (i, ii) PVDF- and (iii) cellulose-based QSPEs, where weak polymer-Li^+ interactions render ion transport predominantly governed by residual solvent (e.g. DMF) coordination [39, 88, 90]; (c) At sufficiently high additive/salt contents, strong coordination between plastic-crystalline or eutectic-forming additives and lithium salts lowers the melting point and induces eutectic melting, giving rise to localized liquid-like regions; (d) Liquid-assisted Li^+ transport in such plastic crystal or eutectic QSPEs, illustrating vehicular diffusion of mobile liquid-like species; (e) Representative plastic-crystalline or eutectic-forming additives (e.g. SN, NMA, and NMU) used in QSPEs, typically combined with LiTFSI to generate highly polar, ion-conductive solvation phases; (f) Mechanism-associated risks arising from mobile liquid-like species in QSPEs, including solvent activity and volatility, and liquid-driven parasitic reactions that form unstable SEI and CEI.

phases (Figure 3e) [32, 41, 100]. Mechanistic investigations of SN-containing poly(ethyl acrylate) (PEA)-based PEs reveal that increasing SN content progressively shifts Li^+ coordination away from the polymer backbone toward additive-rich domains, with long-range ion transport governed predominantly by the mobility of SN-rich regions rather than polymer segmental relaxation [36, 37]. More broadly, such plastic-crystal- or eutectic-derived systems fall within the QSPE category, as long-range Li^+ transport is mediated by confined, non-percolating liquid-like solvation domains instead of polymer-governed coordination pathways.

Owing to their liquid-assisted transport mechanism, QSPEs typically exhibit RT ionic conductivities in the range of 10^{-4}–10^{-3} S cm^{-1}, enabling battery operation without external heating. At the same time, the presence of confined liquid-like domains enhances electrode–electrolyte conformity relative to fully polymer-governed ASPEs, thereby reducing interfacial resistance. These combined attributes position QSPEs as an appealing intermediate platform that balances high ion conductivity of liquid-mediated systems with the mechanical robustness associated with solid-state architectures [101]. Nevertheless, because their ion transport remains liquid-mediated, these QSPEs cannot be regarded as true ASPEs despite their solid-like appearance. Additionally, residual *solvent or liquid-like species in QSPEs may introduce potential safety risks related to solvent activity and volatility, while also participating in interfacial side reactions, contributing to the formation of unstable SEI and CEI, particularly under realistic electrochemical operating conditions* (Figure 3c) [101]. Accordingly, while QSPEs generally offer improved safety and stability relative to GPEs, their absolute safety margins and interfacial robustness remain constrained by the presence of liquid-assisted transport pathways, in contrast to polymer-governed ASPEs. The widespread misclassification of such liquid-assisted systems as truly ASPEs therefore continues to obscure mechanistic understanding, complicate performance benchmarking, and distort assessments of safety and interfacial stability across PE systems [92–94]. This issue is particularly evident when extrapolating results from academic coin cells to multilayer pouch cells, owing to the strong dependence of performance and failure modes on testing conditions. Accordingly, evaluation metrics that reflect practically accessible energy per cycle, incorporating cathode areal loading, operating voltage, and cell configuration, are more meaningful than comparisons based solely on ionic conductivity or specific capacity.

2.3 | All-Solid Polymer Electrolytes (ASPEs)

ASPEs are polymer-based electrolytes in which both ion solvation and transport are governed *exclusively* by the polymer matrix itself, without reliance on liquid-mediated vehicular pathways. In ASPEs, the polymer matrix simultaneously defines the Li^+ coordination environment and the conduction pathway, resulting in transport mechanisms distinct from those in GPEs and QSPEs. In conventional polyether-based systems, Li^+ migration proceeds predominantly via segmental-motion-assisted hopping, where ion transfer between coordinating sites is strongly coupled with local polymer chain relaxation (Figure 4a) [34, 102, 103]. In the absence of liquid-mediated vehicular pathways, long-range ion transport is constrained by Li^+-polymer coordination strength, polymer dynamics, and chain packing [104, 105]. As a result, ASPEs typically exhibit inferior RT ionic conductivities in the range of 10^{-7}–10^{-4} S cm^{-1}, two to four orders of magnitude lower than those of liquid-dominated or liquid-assisted PE systems, rendering stable RT SSBs operation particularly challenging [13, 19, 106, 107].

Despite these transport limitations, the polymer-governed nature of ion transport in ASPEs confers several fundamental advantages, including suppression of evaporation, leakage, and flammability, as well as reduced liquid-driven parasitic reactions at electrode interfaces, thereby enhancing safety and chemical/electrochemical stability [13]. Beyond classical PEO-based electrolytes, alternative polymer hosts, such as polytetrahydrofuran (PTHF), poly(trimethylenecarbonate) (PTMC), poly(ethylene carbonate) (PEC), poly(ε-caprolactone) (PCL), and poly(lactic acid) (PLA) have attracted increasing attention due to their lower crystallinity and weaker Li^+-coordination interactions, which promote chain mobility while preserving polymer-governed ion transport (Figure 4b) [44, 108–114]. More broadly, polymer chemistries afford exceptional molecular and structural design versatility, enabling systematic tuning of functional groups, backbone flexibility, polymer architecture, nanophase morphology, and polymer-ion interactions to optimize local solvation structures, coordination exchange dynamics, and conduction pathways connectivity (Figure 4b) [115–118].

Despite their structural and chemical design versatility, ASPEs face two persistent and interrelated challenges (Figure 4c). The first is their limited RT ion transport capability associated with polymer-governed transport mechanisms. Conventional approaches, such as solid plasticization [22, 42, 30, 119], incorporation of solid-state functional additives (passive or active) [14, 120, 121], copolymerization [122–126], and polymer blending [127–129], primarily operate by lowering the glass transition temperature (T_g), and when applicable, suppressing polymer crystallinity, and accelerating polymer segmental dynamics. However, these strategies remain constrained by the strong coupling between Li^+ transport and polymer segmental dynamics . In the case of inorganic active fillers, when they primarily disrupt polymer crystallinity and enhance amorphous segmental mobility, such that Li^+ transport continues to be polymer-governed, these systems remain within the ASPEs category [130, 131]. In contrast, when sufficiently high filler loadings establish a continuous inorganic conduction network that dominates long-range Li^+ transport and the polymer matrix serves mainly as a mechanical binder, the system transitions toward a hybrid or composite solid electrolyte [132, 133].

One promising direction seeks to weaken Li^+-polymer coupling through site-to-site hopping within transient solvation cages [34, 102], in which weakly coordinated polymers [110, 134, 135], single-ion conducting polymers [136–138], and polymer-in-salt electrolytes [139–142] minimize Li^+-polymer association and allow Li^+ migration via continuous coordination exchange rather than large-scale polymer segmental relaxation (Figure 4d). Alternative emerging strategy relies on topologically engineered ion highways, in which self-assembled or phase-separated ion-rich domains or nanochannels establish continuous, low-tortuosity conduction pathways [129, 143–149]. Importantly, in these systems, ion transport remains polymer-governed,

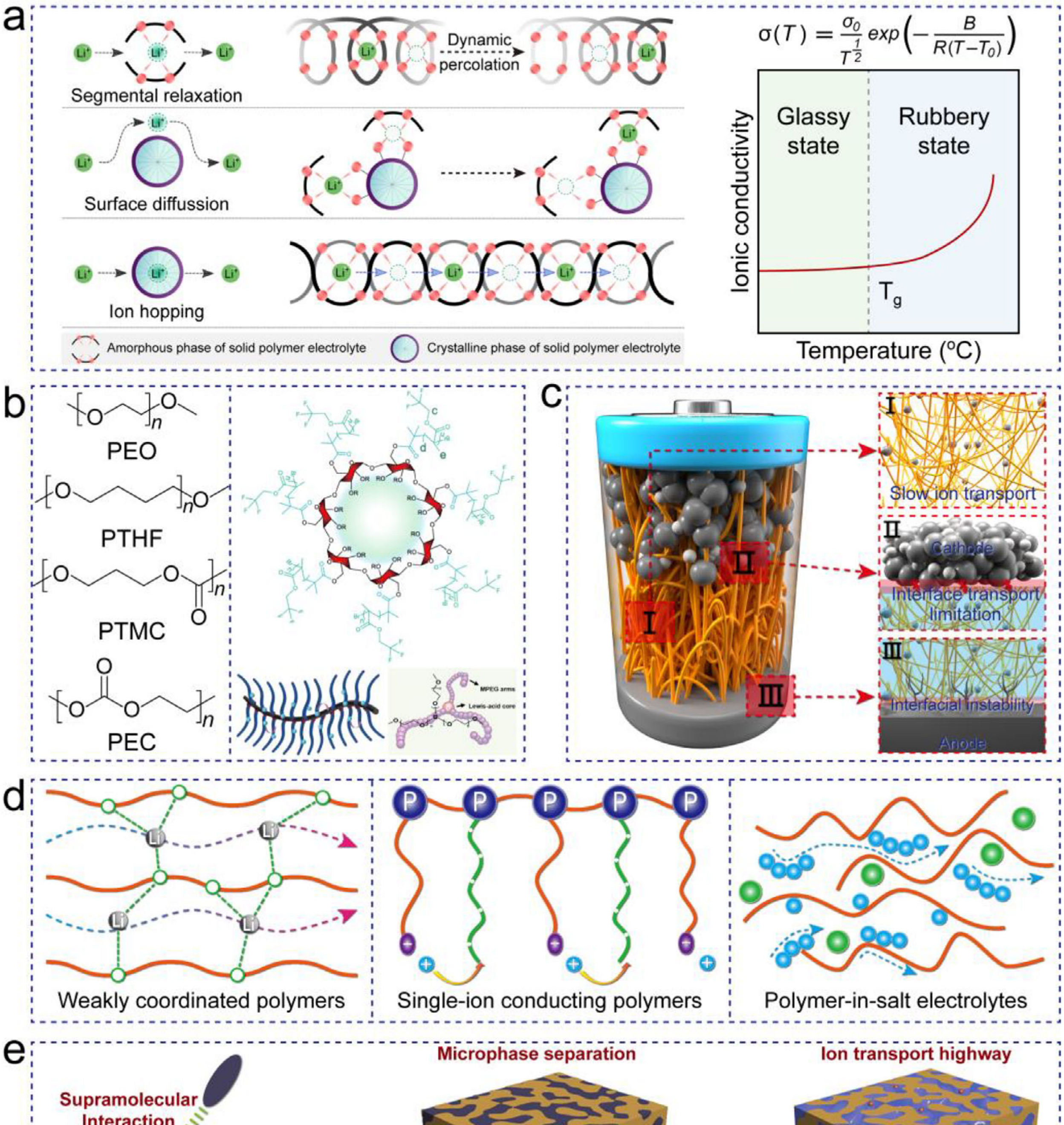

FIGURE 4 | Polymer-governed ion transport and architecture-enabled strategies in ASPEs. (a) Representative polymer-governed Li^+ transport pathways in ASPEs, including segmental-motion-assisted diffusion, interfacial/surface diffusion, and site-to-site ion hopping, together with the temperature-dependent ionic conductivity across the glass transition temperature (T_g) (right panel) [34, 102]; (b) Representative polymer backbones (PEO, PTHF, PTMC, and PEC) and advanced molecular architectures (right panel), illustrating the role of chemical structure and topology in regulating Li^+ coordination and polymer dynamics [44, 108–110, 112, 115–117]; (c) Schematic illustration of bulk and interfacial transport limitations in ASPE-based cells, highlighting restricted ion percolation and solid-solid interfacial issues; (d) Established polymer design strategies to mitigate the coupling between ion transport and polymer segmental relaxation, including weakly coordinated polymers, single-ion conducting polymers, and polymer-in-salt electrolytes; (e) Supramolecular self-assembled polymer architectures that generate ion-rich domains, thereby constructing low-tortuosity, polymer-governed ion-transport highways in ASPEs [147].

while spatial organization of coordinating sites reduces the reliance of long-range transport on bulk segmental motion (Figure 4e). Nevertheless, these transport-enhancement strategies often involve trade-offs between ionic conductivity and mechanical robustness, making it extremely challenging for truly polymer-governed ASPEs to simultaneously achieve RT conductivity above 10^{-4} S cm^{-1} and mechanically stable solid-state architectures suitable for battery operation.

The second major challenge lies at electrode–electrolyte interfaces [15, 62, 150–152]. Unlike liquid-wetted interfaces in GPEs and QSPEs, solid-solid interfacial contact is highly sensitive to mechanical mismatch and electrochemical stress. Poor interfacial wetting, contact loss during cycling, interface thickening, and non-uniform Li^+ flux distribution frequently emerge as dominant failure modes, particularly under high current densities, extended cycling, or limited lithium inventory. Although dynamic bonding chemistries and viscoelastic polymer designs have shown promise in improving interfacial conformity [134, 153–157], maintaining stable operation of SSBs under practical operating conditions (such as high areal capacities, thin electrolyte layer, limited lithium inventory, and elevated current densities) remains a formidable challenge. *Accordingly, interfacial stability in ASPEs should be considered as a dynamic, mechanically coupled process rather than a static material property*. These interfacial limitations are further exacerbated by the tightly coupled chemo-electro-mechanical environment of ASPEs, which imposes far more restrictive constraints than those encountered in liquid-based systems.

3 | Research Priorities of PEs

Given the fundamentally distinct ion-transport mechanisms, structural characteristics, and functional roles of GPEs, QSPEs, and ASPEs, each class faces specific challenges and design constraints that cannot be effectively addressed through a unified research strategy. Accordingly, research priorities should be formulated in a *mechanism-specific and application-relevant manner*, targeting the dominant limitations to each PE class rather than pursuing isolated metrics such as RT ionic conductivity. This section outlines *category-specific research priorities*, with emphasis on *not only ion-transport properties but also interfacial stability, mechanical compatibility, realistic electrode configurations, and cell-level performance metrics* relevant to high-energy-density solid-state batteries. By aligning materials design objectives with the governing ion-transport mechanisms and operational constraints of each PE class, this framework aims to provide a basis for more rational development and more meaningful cross-study comparisons (Figure 5).

3.1 | Gel Polymer Electrolytes (GPEs)

GPEs are among the most accessible PE platforms to achieve high-energy-density battery configurations, owing to their liquid-dominated ion transport pathways and favorable electrode wettability. However, a substantial fraction of reported GPE performance is evaluated under testing conditions that deviate from practical device operation, including low cathode loading, excessive Li inventory, thick electrolyte layers, and low current densities [28, 68, 158, 159]. Such idealized testing protocols obscure the practical limitations of GPE systems and complicate meaningful comparison with state-of-the-art liquid-electrolyte batteries. Accordingly, *a primary research priority for GPEs is to benchmark full-cell performance under application-relevant conditions, including high areal capacity (≥ 3 mAh cm^{-2}), practical N/P ratios (≤ 3), thin electrolyte (≤ 50 μm), and operationally meaningful current densities (≥ 1 mA cm^{-2})* [160–162]. Establishing standardized and application-oriented testing protocols is therefore essential for objectively assessing whether GPEs can genuinely deliver high-energy PE-based battery performance at the full-cell level.

For GPEs, research priorities should shift from maximizing bulk ion transport to improving interfacial stability, mechanical reliability, electrochemical performance, and safety under demanding operating conditions. In this context, the polymer matrix, which distinguishes GPEs from conventional liquid electrolytes, should be regarded as a central design variable rather than serving merely as a mechanical support. Rationally engineered polymer networks provide mechanical confinement and spatial regulation of the liquid electrolyte, suppressing uncontrolled electrolyte redistribution during fast charging or prolonged cycling [59, 101, 163]. The viscoelastic nature of polymer networks further enables accommodation of electrode volume changes, helping to preserve interfacial contact and mechanical integrity throughout cycling [28, 164]. As a result of regulated electrolyte distribution and buffered chemo-mechanical response, the well-designed polymer networks can suppress lithium dendrite penetration and delay mechanical failure under aggressive operating conditions. Beyond structural reinforcement, advanced polymer network designs further enable system-level functionalities that are difficult to realize in purely liquid electrolytes, including flame retardancy, thermo-responsive shutdown behavior, autonomous or damage-tolerant self-healing, and improved recyclability [165–169]. Collectively, these attributes highlight the role of polymer networks in GPEs as active enablers of safety, reliability, and long-term durability rather than passive mechanical supports.

Beyond mechanical regulation and functionality, further advancement of GPEs increasingly relies on deliberate polymer-solvent-salt co-design to control ion solvation and interfacial chemistry under confinement. Although solvent properties (e.g. donor number, polarity, viscosity, oxidative stability, and volatility) directly influence Li^+ coordination, mobility, and chemical reactivity, polymer-solvent interactions dictate solvent uptake, confinement, solvation structures, local activity, and spatial distribution within the gel matrix [170–172]. Polymer-ion interactions regulate anion motion, local concentration heterogeneities, and interface reactivity, collectively shaping the effective Li^+ solvation environment during operation [173–176]. Poor compatibility between polymer, solvent, and electrolyte salts leads to phase separation, enhanced solvent activity, and unstable SEI/CEI formation, accelerating parasitic reactions and compromising electrochemical stability. Conversely, synergistically designed polymer-solvent-salt systems can stabilize favorable Li^+ solvation motifs, suppress liquid-driven side reactions, and regulate interfacial chemistry under dynamic operating conditions [34, 66, 105, 161, 173]. *Future research should therefore focus on (i) identifying solvent-salt combinations that retain stable coordination structures under polymer confinement*

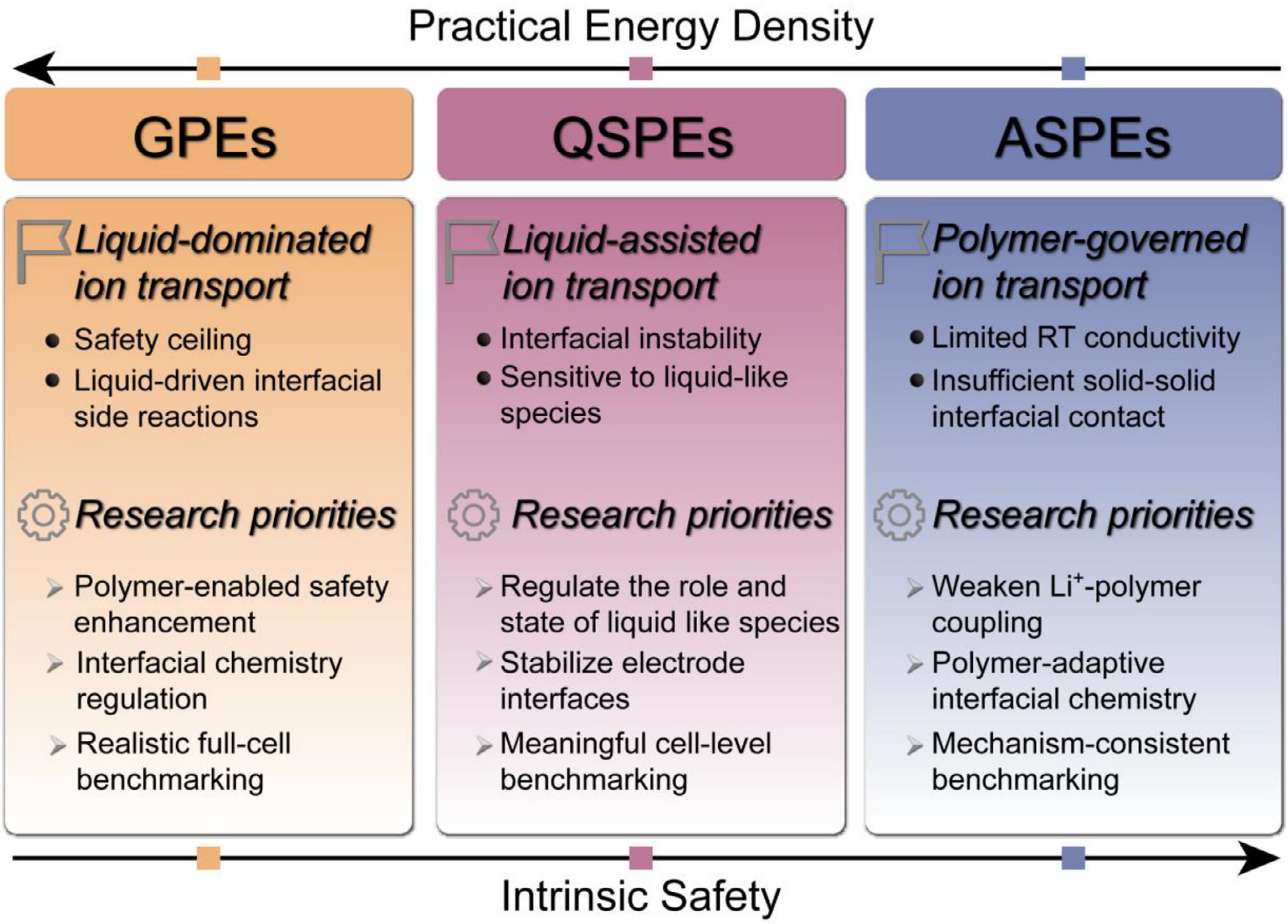

FIGURE 5 | Mechanism-derived research priorities across PE systems. Schematic summary of category-specific research priorities for GPEs, QSPEs, and ASPEs, derived from their distinct ion-transport mechanisms and structural constraints along the axes of practical energy density and safety. For GPEs, priorities center on polymer-enabled safety enhancement, interfacial regulation under realistic conditions, and full-cell-level benchmarking to address safety limitations associated with liquid-dominated transport. For QSPEs, key priorities focus on controlling and quantifying liquid-like species, stabilizing interfaces at both lithium metal anode and high-loading composite cathodes, and establishing meaningful cell-level benchmarking. For ASPEs, limited RT ionic conductivity and solid-solid interface contact define the main bottlenecks, motivating strategies to mitigate the dependence of ion transport on polymer segmental dynamics, stabilize solid-solid interfaces through polymer-adaptive interfacial chemistry, and develop mechanism-aligned benchmarking frameworks.

and (ii) engineering polymer matrices capable of selectively tuning solvent activity and Li$^+$ solvation to enhance interfacial stability and safety. Shifting the design focus from maximizing ionic conductivity to regulating solvation, interfacial chemistry through polymer design enables GPEs to evolve into functionally integrated hybrid systems with enhanced safety, durability, and electrochemical performance [168].

3.2 | Quasi-Solid Polymer Electrolytes (QSPEs)

QSPEs occupy an intermediate regime in which electrochemical performance is governed by the presence and behavior of confined liquid-like species within the polymer matrix, rather than by the polymer framework alone. These species play a central role in both Li$^+$ transport and interfacial chemistry, enabling decent RT ionic conductivity and favorable initial electrode wettability. However, because ion transport and interfacial chemistry are strongly coupled to the content, spatial distribution, and electrochemical activity of these liquid-like species, the performance of QSPEs remains sensitive to *formulation-dependent solvation structures and their evolution* under electrochemical cycling processes [173, 177, 178]. Research on QSPEs should therefore focus on quantifying and regulating the content, spatial distribution, and electrochemical activity of confined liquid-like species during operation.

A central priority for QSPE-based batteries is to mitigate liquid-driven interphase instability, particularly at the lithium metal anode [141, 174, 179]. Residual solvents or liquid-like species can continuously participate in electrochemical reactions during cycling, leading to chemically unstable and progressively thickening interphases, parasitic reactions, and deterioration of Coulombic efficiency (CE). These effects become more pronounced under practical operating conditions, such as increased current densities, extended cycling, and limited lithium inventory, where solvent-driven side reactions rapidly accumulate. Similar constraints arise at the composite cathode side, where liquid-assisted transport can impede uniform ion penetration across thick, high-mass-loading electrodes, thereby limiting achievable areal capacity and rate capability [43, 180–182]. Moreover, solvent molecules and solvated species infiltrating porous cathode architectures are increasingly susceptible to electrochemical oxidation under high-voltage operation, further exacerbating CEI instability. Consequently, *performance optimization strategies that rely primarily on improved ionic conductivity via additional liquid components often deliver short-term gains at the expense of long-term interfacial stability, voltage tolerance, and safety*. Future research should therefore prioritize solvation and liquid components management, including minimizing electrochemically active "free" liquid-like species, confining solvation structures through polymer coordination and high-salt environments, and stabilizing interphases via robust SEI/CEI chemistries rather than pursuing ASPE-like claims based solely on ionic conductivity.

Equally important is the evaluation and reporting framework used for QSPEs. In liquid-assisted systems, standalone metrics such as bulk ionic conductivity or linear sweep voltammetry (LSV) can be misleading: high ionic conductivity simply reflect liquid-assisted ion transport, while apparent oxidative stability may not capture solvent infiltration into composite cathodes or time-dependent interphase growth. *Meaningful benchmarking of QSPEs therefore requires: transparent quantification of liquid-like content and its state (free vs confined) using complementary methods (e.g. NMR, TGA-MS, or Karl Fischer titration)* [183], *standardized CE evaluation (Aurbach CE protocols)* [182] and *interphase-sensitive diagnostics under lean-lithium and practical current densities, quantification of oxidative side reactions under relevant potentials and time scales (floating analysis)* [75, 177, 178, 184] *and cell-level testing with realistic cathode loadings, practical N/P ratios, and thin electrolyte layers*. These requirements are particularly critical for QSPEs, as their solid-like appearance can obscure the mechanistic role of confined liquid-like species unless composition and state are explicitly reported. From a safety perspective, simple ignition tests are insufficient; instead, comprehensive thermal-abuse and mechanical-abuse evaluation, including accelerating aging, self-heating kinetics, and full-cell abuse testing, is needed to support safety claims for QSPEs [167, 181]. Taken together, research on QSPEs should focus on *controlling the presence, distribution, and interfacial reactivity of liquid-like species*, supported by *transparent quantification methods and application-relevant benchmarking protocols*. This mechanism-aligned framework clarifies the limitations of evaluating QSPEs using ASPE-derived metrics alone and delineates practical pathways toward improved interfacial stability, enhanced safety, and robust full-cell performance in near-solid-state battery configurations.

3.3 | All-Solid Polymer Electrolytes (ASPEs)

Among PE systems, ASPEs represent the most stringent platform for pursuing safe, high-energy-density solid-state batteries because both transport and interfacial behavior must be realized without liquid-mediated assistance. Despite sustained research efforts, the electrochemical performance of ASPE-based batteries remains substantially inferior to that of GPE- and QSPE-based counterparts, particularly under RT and high-energy-density operating conditions. Currently, the practical operation of ASPE systems often relies on elevated temperatures and remains challenging under realistic cell configurations [117, 118, 134]. Therefore, research efforts on ASPEs should be redirected toward addressing their underlying transport and interfacial limitations, rather than relying on apparent performance gains derived from liquid-mediated mechanisms.

A central research direction for ASPEs is to enhance ionic conductivity while preserving a strictly polymer-governed conduction mechanism. In practice, *a considerable number of PEs reported "all-solid" still rely on trace amounts of liquid-like species, such as residual solvents, unreacted monomers, eutectic phases, or additional interface liquids (such as fluoroethylene carbonate (FEC)), that actively assist ion transport and electrode–electrolyte contact, thereby inflating apparent ionic conductivity and electrochemical performance* [48, 117, 185, 186]. While such strategies can improve measured ion transport properties and interfacial conformity under idealized conditions, *they alter the dominant ion-transport mechanism by introducing liquid-mediated pathways that deviate from truly polymer-governed conduction*. The resulting performance metrics often exceed those achievable in *strictly polymer-governed* ASPEs. Such mechanistic inconsistency complicates a fair comparison across ASPE studies and obscures fundamental polymer structure-property relationships. Therefore, confirming polymer-governed transport and transparently reporting any liquid-like content and state should be treated as a prerequisite for meaningful ASPE benchmarking.

Enhancing RT ionic conductivity remains necessary for ASPE operation through the design of the polymer matrix and electrolyte salt. Polymer-governed ion transport is restricted by coupled thermodynamic and kinetic constraints, involving the dissociation of electrolyte salt, Li^+-polymer coordination dynamics, and segmental mobility of the polymer matrix. Increasing polymer polarity or the density of coordinating sites generally promotes salt dissociation, thereby increasing the concentration of charge carriers. However, stronger Li^+-polymer interactions simultaneously raise the energetic barrier for coordination exchange and can retard local segmental dynamics, leading to reduced ion mobility. Conversely, weakening Li^+ coordination facilitates faster hopping and segmental motion but often results in insufficient salt dissociation and a lower fraction of mobile ions [187]. Consequently, addressing this trade-off *requires a mechanism-resolved understanding of ion dissociation, coordination exchange, and migration pathways, supported by quantitative descriptors such as Li^+ coordination environment, Li^+ transference number, and temperature-dependent transport behavior*. Additionally, improvement in bulk ion-transport properties must be closely correlated with mechanical integrity, interfacial stability, and electrochemical durability, ensuring sustained lithium stripping/plating behavior under practical operation conditions. Notably, reported battery performance should be interpreted cautiously, as they are strongly influenced by cell configuration and testing conditions rather than arising solely from electrolyte characteristics [188].

Beyond improving bulk ion transport, ASPE research should develop intimate interfaces that can maintain contact, accommodate stress, and evolve stable interphases without liquid-mediated self-adjustment. In contrast to liquid-mediated GPEs and QSPEs, ASPEs depend on the viscoelastic response, interfacial adhesion, and chemical adaptability of polymer matrix to maintain intimate interfacial contact while accommodating volume changes associated with lithium metal anodes and composite cathodes during lithium plating/stripping processes. Interfacial contact loss, stress accumulation, internal cell pressure, interphase thickening, and non-uniform Li^+ flux distribution therefore emerge as critical failure modes, particularly under high current density and extended cycling [15, 151, 156, 189]. Progress in ASPEs therefore critically depends on integrating polymer design with adaptive interfacial chemistry that enables dynamic stress accommodation and stable interphase evolution without introducing liquid-mediated pathways.

Given the intrinsic limitations of polymer-governed ion transport, benchmarking ASPEs requires a staged and mechanism-aligned framework rather than direct comparison with liquid-dominated or liquid-assisted systems under identical conditions. Ionic conductivity target alone, such as exceeding 10^{-4} S cm^{-1}, does not guarantee stable operation under practical current densities or high areal capacities, nor do they adequately reflect the underlying constraints of polymer-governed ASPEs. *Meaningful evaluation should therefore prioritize confirmation of polymer-governed transport mechanisms, sustained lithium stripping/plating behavior, and long-term interfacial and mechanical stability under moderate yet electrochemically relevant conditions*. Accordingly, standardized and application-relevant benchmarking protocols are essential to avoid unrealistic expectations, enable meaningful comparison across ASPE systems, and provide a transparent pathway for assessing genuine advances in polymer electrolyte design. Despite these persistent and formidable challenges, continued advances in polymer chemistry, electrochemistry, interfacial science, and battery technology are expected to progressively unlock the potential of ASPEs, ultimately accelerating progress toward practical SSBs applications.

4 | Conclusions and Outlook

GPEs, QSPEs, and ASPEs constitute three mechanistically distinct classes of PEs, differentiated by their dominant long-range Li^+ transport pathways, ranging from liquid-dominated, through liquid-assisted transport to fully polymer-governed ion conduction, rather than by macroscopic appearance or composition alone. These mechanistic distinctions give rise to systematic differences in ionic conductivity, interfacial behavior, mechanical response, safety characteristics, and cell-level performance (Figure 6a), as further summarized in Table 1 in terms of representative mechanistic features, electrochemical performance, and practical evaluation metrics for each PE class.

GPEs benefit from liquid-dominated ion transport and excellent electrode wettability, enabling high RT ionic conductivity and low interfacial resistance; however, the presence of a percolating liquid phase introduces safety and long-term reliability concerns, including volatility, leakage, flammability, and parasitic interfacial reactions. QSPEs occupy an intermediate regime, where ion transport is enabled by mobile liquid-like species confined within a solid-like polymer matrix, thereby offering a balance between ionic conductivity and mechanical robustness. Nevertheless, their practical advancement toward high-energy-density and safe batteries remains limited by liquid-related potential risks, interfacial instability with Li metal anode, and insufficient Li^+ transport across thick, high-mass-loading composite cathodes under practical operating conditions. In contrast, ASPEs rely exclusively on polymer-governed ion transport and eliminate liquid-mediated pathways, positioning them as the most stringent route toward intrinsically safe ASSBs. Their practical implementation, however, remains hindered by sluggish polymer-governed ion transport kinetics and insufficient solid-solid interfacial contact, particularly under RT and realistic operating conditions involving high areal current/capacity, limited lithium inventory, and high cathode loading. Collectively, these characteristics underscore that each PE class reflects a unique balance among ionic conductivity, mechanical integrity, interfacial stability, and safety, and that no single strategy can be universally applied across all PE systems.

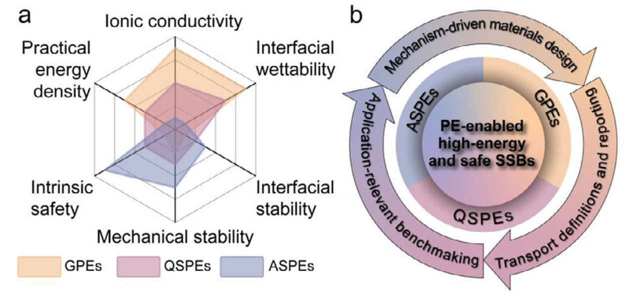

FIGURE 6 | Mechanistic comparison and unifying framework of PEs. (a) Qualitative radar comparison of GPEs, QSPEs, and ASPEs across six key performance dimensions, that is, ionic conductivity, interfacial wettability, interfacial stability, mechanical stability, intrinsic safety, and practical energy density, illustrating intrinsic trade-offs dictated by their distinct ion-transport mechanisms rather than absolute performance metrics; (b) A unified conceptual framework for accelerating PE-enabled high-energy-density and safe SSBs, emphasizing the iterative integration of mechanism-based transport definitions and standardized reporting, mechanism-driven materials design, and application-relevant benchmarking.

Advancing PE-enabled high-energy-density and safe batteries therefore requires category-specific strategies grounded in *mechanism-driven materials design, transparent definitions and reporting, and application-relevant benchmarking* (Figure 6b). For GPEs, the focus should shift from maximizing ionic conductivity toward demonstrating safety, durability, and energy density comparable to state-of-the-art liquid electrolyte systems under realistic full-cell conditions. For QSPEs, progress hinges on regulating and explicitly reporting the role and physicochemical state of liquid-like species, ensuring effective ion transport while suppressing persistent interfacial instability and liquid-associated safety risks, and avoiding their misclassification as all-solid systems. For ASPEs, research should prioritize enhancing RT polymer-governed ion transport and engineering intimate, conformal solid-solid interfaces to advance their practical application, while rigorously avoiding liquid-mediated pathways that obscure mechanistic interpretation and exaggerate performance claims.

Looking forward, the transition of PEs from laboratory demonstrations to commercially relevant technologies will depend

TABLE 1 | Representative mechanistic features, electrochemical performance, and practical evaluation metrics for each PE class.

PE class	Composition and mechanistic features	Electrochemical performance			Mechanical properties	Interface resistance (Ω)	Practical battery metrics			Refs.
		Ionic conductivity (S cm^{-1})	Li^+ transference number	Electrochemical stability window (V)			Areal current/capacity (mA cm^{-2}/mAh cm^{-2})	Cathode	Pouch cell or Energy density	
GPEs	1 M LiTFSI in DOL/MP/FEC (4:5:1) with 3 wt.% LiDFOB MP/FEC-dominated transport	9.40×10^{-3} (30°C)	0.84	5.2	/	<300	1/1	NMC811: 1.2 mAh cm^{-2}	3 mAh cm^{-2}	[160]
	TF + HCE (5.2 M LiFSI/DME) DME-dominated transport	4.02×10^{-3} (25°C)	0.63	5.3	/	<50	1/2	NCM811: 2.09 mAh cm^{-2}	465.63 Wh kg^{-1}	[161]
	PVM(1 M LiBOB+0.2 M $LiNO_3$ in PC/DME) PC/DME-dominated transport	1.4×10^{-3} (30°C)	0.77	4.74	/	200–350	1/1; 3/1.5	NMC811: 3 and 8 mg cm^{-2}	1 Ah	[162]

(Continues)

TABLE 1 | (Continued)

PE class	Composition and mechanistic features	Electrochemical performance			Mechanical properties	Interface resistance (Ω)	Practical battery metrics			Refs.
		Ionic conductivity (S cm^{-1})	Li$^+$ transference number	Electrochemical stability window (V)			Areal current/capacity (mA cm^{-2}/mAh cm^{-2})	Cathode	Pouch cell or Energy density	
QSPEs	PVDF + LiFSI + 15%DMF DMF-Li$^+$-assisted transport	3×10^{-4} (RT)	0.44	~4.8	Young's modulus: 45.87 MPa	<200	0.2/0.2	NMC111: 2.5 and 12 mg cm^{-2}	/	[39]
	PEA + 30%LiTFSI + 50%SN + 5%FEC SN-LiTFSI-assisted transport	1.01×10^{-3} (RT)	/	4.9	/	/	0.5/1	LFP: 3 mg cm^{-2}	Demo	[37]
	PTF-PE + 30% TMP TMP-Li$^+$-assisted transport	6.6×10^{-4} (25°C)	/	~5.1	Stress: 0.1 MPa	~200	0.2/0.2; 0.2/8	LRMO:3.5 mg cm^{-2}	600 Wh kg^{-1}	[178]
ASPEs	HEMI + 12%LiTFSI	2.66×10^{-6} (30°C)	/	/	Stress: 8.7 MPa	/	/	/	/	[118]
	PEO-governed transport	4.56×10^{-4} (70°C)	0.63	4.28	/	800–1000	0.1/0.1	LFP: 1–2 mg cm^{-2}		
	alterSIPE, P8 Single-ion conductor	4.0×10^{-5} (30°C)	0.93	/	/	/	0.15/0.15	LFP: 8 mg cm^{-2}	/	[138]
	PEO + MBO + LiTFSI PEO-governed transport	~10^{-5} (30 C)	/	/	Stress: 4.13 MPa Young's modulus: 1.84 MPa	/	/	LFP/NMC811:1.0–1.2 mg cm^{-2}	Demo (12.4 mg cm^{-2} LFP)	[117]
		2.7×10^{-4} (50°C)	0.63	5.08		400–800	0.05/0.05			

less on isolated performance metrics and more on mechanism-centered materials innovation, supported by standardized characterization and application-relevant testing protocols. Establishing a unified mechanistic understanding, transparent benchmarking and reporting, and clearly defined failure criteria across PE systems will be critical for enabling meaningful comparison and rational materials selection. By grounding electrolyte development in rigorous methodology, mechanistic understanding, and molecular-level design innovation, the PEs community will be positioned to deliver next-generation SSBs that combine high-energy-density, long-term durability, and intrinsic safety, ultimately enabling PEs to compete with both liquid and inorganic solid-state electrolyte technologies.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Research data are not shared.

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