Research Progress on Composite Solid Polymer Electrolytes for All-Solid-State Batteries

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Abstract. The growing demand for high-energy-density, long-life and safe energy-storage systems has accelerated the development of all-solid-state batteries (ASSBs), particularly for electric vehicles and grid-scale storage. Conventional liquid electrolytes suffer from flammability, leakage and thermal runaway risks and are increasingly unable to meet the requirements of next-generation batteries. Composite solid polymer electrolytes (CSPEs), which combine polymer matrices with inorganic fillers, offer a promising alternative. Compared with inorganic solid electrolytes (ISEs) and pure solid polymer electrolytes (SPEs), CSPEs provide a more balanced combination of flexibility, processability, ionic conductivity, mechanical robustness and interfacial compatibility. This review systematically summarizes the fundamental components including polymer matrices, inorganic fillers and lithium salts and ion-conduction mechanisms. Although recent studies have elucidated various ion-conduction mechanisms within CSPEs, these mechanisms remain to be further explored and understood at a deeper level. Various design and modification strategies are discussed in detail, with particular attention given to the interfacial issues between CSPEs and electrodes as well as the corresponding optimization strategies. Finally, the remaining challenges are also discussed, providing insights for future development of high-performance CSPEs for practical ASSB applications.

Keywords: All-Solid-State Batteries (ASSBs); Composite Solid Polymer Electrolytes (CSPEs); Interface Engineering; Modification Strategy.

1. Introduction

The market demand for energy storage systems with high energy density, high safety and long cycle life for electric vehicles, portable electronics and large-scale grid storage is booming. It leads to a rapid progression of next-generation rechargeable battery technologies. Conventional liquid-electrolyte lithium-ion batteries (LIBs) have been the dominant technology in these fields because of its mature manufacturing infrastructure and good energy density. However, they have now reached their energy density limit and serious safety risks like flammability, electrolyte leakage and thermal runaway also exist. Thus, the problems prohibit its application in novel high-performance applications. For settling these safety issues, all-solid-state batteries (ASSBs) replace non-flammable solid electrolytes for the combustible organic liquid electrolytes to eliminate safety risks. Moreover, ASSBs are well embedded with high-performance electrode materials like lithium metal anodes, which can remarkably increase the cell-level energy density [1,2].

In ASSBs, solid electrolytes serve as pivotal components to control the ion transport, prevent the electron leakage and maintain structural integrity. These functional materials fundamentally determine the comprehensive performance and safety of the battery system [3]. Figure 1 summarizes the key performance characters of prevailing electrolyte categories, which are currently divided into inorganic solid electrolytes (ISEs), pure solid polymer electrolytes (SPEs) and composite solid polymer electrolytes (CSPEs) [4]. This category exhibits that ISEs claim to extra ionic conductivity with a great mechanical rigidity despite their inherent brittleness, poor electrode–electrolyte interfacial compatibility and high manufacturing complexity restricting the commercialization, whereas SPEs (PEO, PVDF/HFP and PMMA) provide great flexibility, good process ability, favorable interfacial wettability with inferior ionic conductivity and limited electrochemical stability

window (ESW), insufficient mechanical strength which greatly restricts its practical realization in high performance ASSBs [2,4].

CSPEs could be achieved by integrating inorganic fillers into polymeric matrices, that combine the complementary advantages of polymeric matrices and inorganic fillers and overcome their respective disadvantages [2]. Concretely, the polymer matrix delivers favorable flexibility and processability, whereas inorganic fillers efficiently reduce polymer crystallinity, enhance ionic conductivity, reinforce mechanical robustness and improve interfacial compatibility with electrodes [3]. Substantial progress has been achieved in CSPEs research and numerous modification strategies have been established to enhance their comprehensive electrochemical properties. Huang et al. demonstrated that a fluoropolyether-based quasi-solid polymer electrolyte (FPE-SPE) can construct anion-derived fluorine-rich interphases to stabilize the LRMO cathode and suppress oxygen release, enabling anode-free pouch cells with a remarkable energy density of 604 Wh kg^{-1}, long cycling stability over 500 cycles, and excellent safety under nail penetration and high-temperature tests [5]. In Li-S batteries, CSPEs can effectively suppress the polysulfide shuttle effect, thereby enabling high reversible capacity and excellent rate capability in all-solid-state Li-S cells [6]. Importantly, CSPEs display outstanding practical applicability and have been widely investigated in versatile ASSB configurations, demonstrating strong feasibility and commercialization promise [4].

Benefiting from the intrinsic flexibility and desirable processability of CSPEs, CSPE-supported flexible ASSBs exhibit outstanding mechanical stability and operational reliability, thereby showing great promise wearable electronics, foldable devices and flexible energy-storage systems for next-generation. Yu et al. constructed a flexible ASSB using PVDF-HFP-based composite polymer electrolytes combined with freestanding carbon nanotube-supported phosphate electrodes, which enables ambient-atmosphere assembly and stable operation under repeated bending and folding [7]. Panneerselvam et al. further fabricated flexible electrospun nanocomposite polymer electrolytes by incorporating NASICON-type $Li_{1.45}Al_{0.45}Ge_{0.2}Ti_{1.35}(PO_4)_3$ (LAGTP) into PVdF-HFP matrices, delivering high ionic conductivity, favorable mechanical deformability and a wide ESW up to 5 V [1].

Scalable fabrication strategies for CSPEs have been extensively explored, including solution casting, hot pressing and in situ polymerization. Solution casting offers low-cost and large-area membrane manufacturing with precise thickness control, while in situ polymerization significantly improves interfacial contact and simplifies cell assembly. However, challenges still exist in cost control, batch-to-batch consistency and scalable production of defect-free thin membranes (20–50 μm). Recent progress has enabled CSPE-based all-solid-state batteries to deliver gravimetric energy densities exceeding 400 Wh/kg and stable cycling over 1000 cycles with 80% capacity retention, typically based on PEO-derived in situ polymerized systems and sulfide/polymer composite electrolytes coupled with high-loading NCM811 cathodes [8-10]. While great improvements have been achieved, CSPEs are still restricted from practical application due to poor ionic conductivity at room temperature (RT), weak electrode-electrolyte interfacial stability, inadequate dendrite resistance and difficulties in scalable, low-cost production with high uniformity.

This review concentrates on the research progress of CSPEs for ASSBs, systematically summarizes the composition, structure and ion transport mechanisms of CSPEs, elaborates on their design and modification strategies and analyzes key electrode-electrolyte interfacial issues alongside corresponding interface optimization strategies. Finally, the main bottlenecks and potential future research orientations of CSPEs are prospected. It is expected to provide in-depth references for researchers and to advance the development and commercialization of CSPE-based ASSBs.

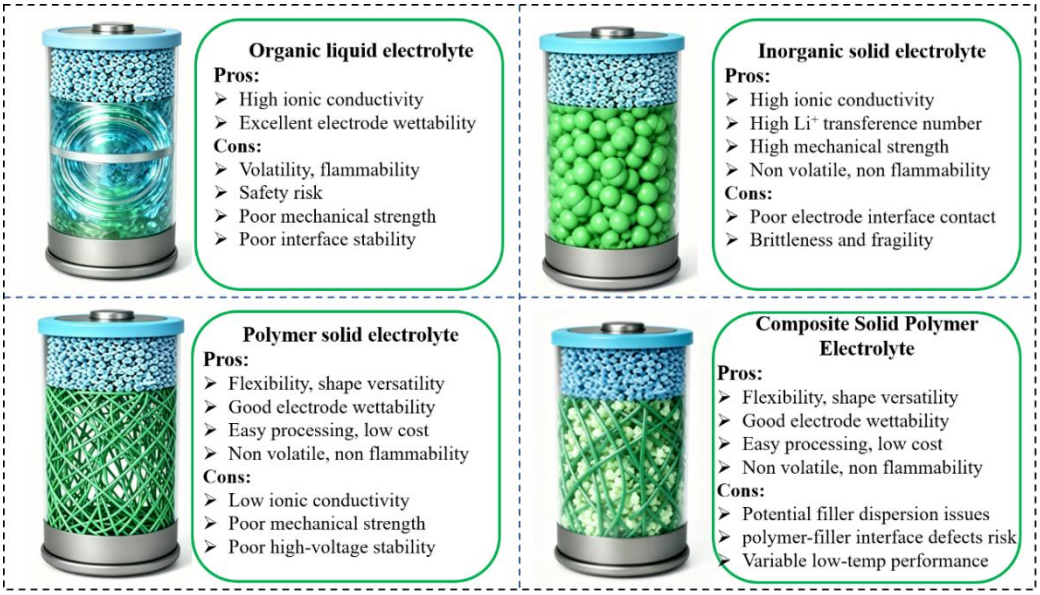

Fig 1. Performance comparison of different electrolytes (Picture credit: Original).

2. Composition of CSPEs

The polymer matrix is the core component of CSPEs, providing a flexible framework for ion conduction and determining the basic properties of CSPEs such as flexibility, processability and interfacial compatibility. Figure 2 illustrates the generalized structure of polymer-based ASSBs and summarizes representative anode, cathode and electrolyte materials that govern cell energy density and power density performance. The ideal polymer matrix for CSPEs should have the following characteristics: good solubility for lithium salts, high electrochemical stability, low crystallinity (to facilitate ion conduction), excellent compatibility and flexibility with inorganic fillers and electrodes. It visually shows how CSPEs and ISEs are integrated into the battery structure, along with typical materials for each component. At present, polyethylene oxide (PEO), polyvinylidene fluoride-hexafluoropropylene (PVDF/HFP), polymethyl methacrylate (PMMA), polyacrylonitrile (PAN), polycarbonate (PC) and polysiloxane are the commonly used polymer matrices in CSPEs.

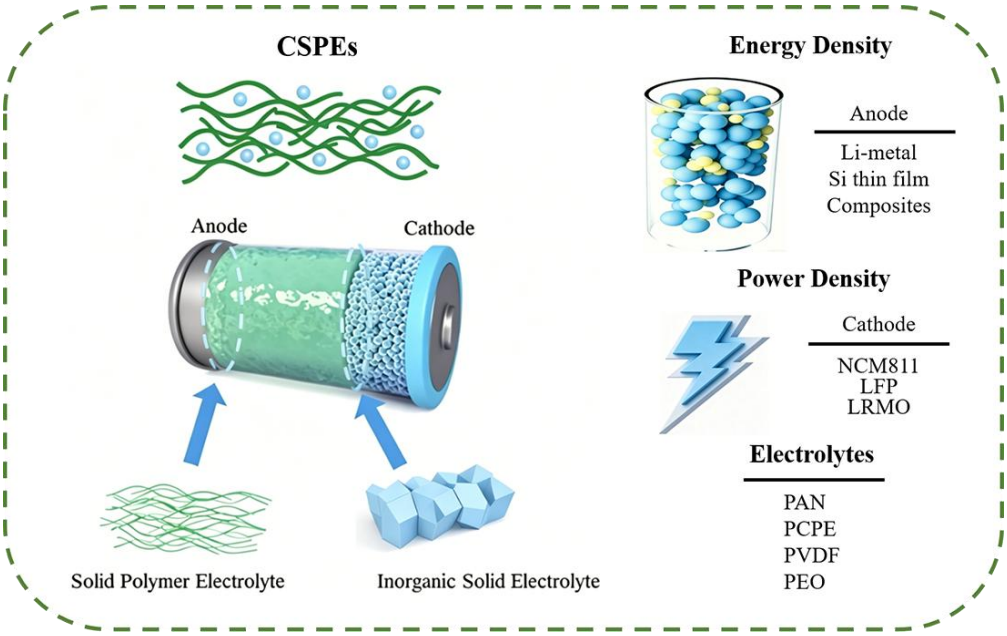

Fig 2. Schematic diagram of a composite solid polymer electrolyte (CSPE)-based cell configuration, illustrating the materials for the anode, solid polymer electrolyte (SPE) and cathode (Picture credit: Original).

2.1. Polymer Matrix

From the perspective of Li^+ migration mechanism, polymer materials used as CSPEs are required to fulfill two essential prerequisites for efficient Li^+ conduction. First, the polymer matrix should contain abundant polar functional groups, which can effectively solvate lithium salts, promote the full dissociation of Li^+ from lithium salt molecules and provide sufficient charge carriers for ion transport within the electrolyte system. Second, the polymer backbone ought to possess appropriate free volume and favorable chain segment mobility, enabling Li^+ to achieve continuous migration via coordination interaction and ion hopping behavior among polymer segments.

In CSPEs studies, PEO remains the dominant polymer matrix, primarily because it can effectively solvate Li^+ through ether oxygen sites, offers good mechanical flexibility and is easy to fabricate. However, at room temperature, PEO's high crystallinity restricts polymer segment motion, leading to poor ionic conductivity (usually 10^{-8}–10^{-6} S/cm at 25 ℃). PVDF-HFP is characterized by high electrochemical stability (with an electrochemical window reaching 5.0 V vs. Li/Li^+), favorable mechanical strength, and low crystallinity. but its interfacial compatibility with lithium metal anodes is poor and it is easy to react with lithium metal to form unstable solid electrolyte interphase (SEI) films. PMMA has good transparency, low crystallinity and excellent compatibility with inorganic fillers, but its mechanical strength is poor, and its electrochemical stability needs to be further improved. PAN's high polarity enables it to greatly enhance the dissociation of lithium salts, but its flexibility is poor and it is easy to decompose at high temperatures. PC and polysiloxane have the advantages of low glass transition temperature (Tg) and good flexibility and are often used as modified components to enhance low-temperature performance and flexibility of CSPEs [10].

2.2. Inorganic Fillers

Inorganic fillers serve as critical functional components in CSPEs. They effectively enhance the ion transport properties, mechanical robustness and interfacial behavior of CSPEs through three key mechanisms: modulating the crystallinity of the polymer matrix, constructing extra pathways for ion migration and optimizing the interfacial contact [8]. Based on their functional characteristics, these fillers are categorized into inert ceramics, Li^+-conductive phases and advanced functional fillers.

Inert ceramic fillers lack intrinsic Li^+ conduction capability but can suppress the crystalline behavior of the polymer matrix, expand the free volume of polymer segments and improve both the mechanical strength and interfacial wettability of CSPEs. For example, tantalum-doped lithium lanthanum zirconium oxide (LLZTO) as an active filler can effectively improve the ionic conductivity and interfacial stability of cross-linked CSPEs. Novel functional fillers (such as MOFs, COFs, GO, h-BN and two-dimensional transition metal sulfides) have multiple functions such as inhibiting lithium dendrite growth, selective ion transport and improving interfacial stability. For example, two dimensional MOFs (Cu-BTC) with unsaturated metal site anchor TFSI to release Li^+ to build multiple transport channels which improve Li^+ transference number and ionic conductivity for CSPEs. Moreover, the crystal facets of inorganic fillers have also an impact on the performance of CSPEs. The facets of (001) BiOCl nanosheets show excellent LiTFSI dissociation capability and mobility in Li^+ compared with (010) facets [8].

2.3. Lithium Salts

Lithium salts are the primary source of Li^+ for CSPEs, this means that the dissociation behaviors and solubility of these salts in polymer matrix directly controls the ionic conductivity and lithium-ion transference number of electrolyte system [10]. To achieve efficient ion transport, an ideal lithium salt should have high dissociation degree, good solubility within the polymer host and a good ESW as well as strict chemical inertness to the polymer matrix and inorganic fillers to suppress parasitic side reactions. These strict performance requirements have caused contemporary research mainly to use the LiTFSI, lithium hexafluorophosphate ($LiPF_6$), and lithium perchlorate ($LiClO_4$) as the standard electrolyte solutes in CSPEs.

LiTFSI is the most used lithium salt in CSPEs with a high dissociation degree, good solubility in majority of polymer matrices and high electrochemical stability. Specifically, this salt has good dissociation efficiency in PEO based CSPE, the detached Li^+ ions can form the coordination bond with ether oxygen group within the PEO to realize the ion conduction. In contrast, $LiPF_6$ easily decomposes in the presence of water, its high ionic conductivity directly deteriorates the whole stability of CSPEs. Also, although the good solubility of $LiCLO_4$ in the polymer matrices, the inferior ESW highly induces that the compound is highly likely to decompose at the high voltage, which strictly restricts its application in the high voltage ASSBs [10]. Sulfide type fillers such as $Li_{10}GeP_2S_{12}$ and Li_6PS_5 presented excellent ionic conductivity as evidenced by PEO-based CSPEs containing 97 wt% Li_6PS_5Cl have a conductivity of 1.1×10^{-3} $S \cdot cm^{-1}$ and a wide ESW of 4.9 V. To improve both ion transport and ESW simultaneously, the researchers had frequently introduced the garnet type fillers including $Li_7La_3Zr_2O_{12}$ and $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ into polymer host.

3. Ion Conduction Mechanism in CSPEs

The boost of ionic conductivity of CSPEs compared to the single phase counterparts is from a synergic transport mechanism combining polymer host relaxation, lithium-ion migration and filler driven interfacial effects (Figure 3b). As characterized in Figure 3a, the conventional transport mode in unreinforced polymers depend on the segmental relaxation and local surface diffusion intensely. The incorporation of inorganic fillers substantially modified the underlying dynamics. These functional additives inhibit polymer crystallization and enlarge the local amorphous domains, which establish the continuous and low resistance conduction channels.

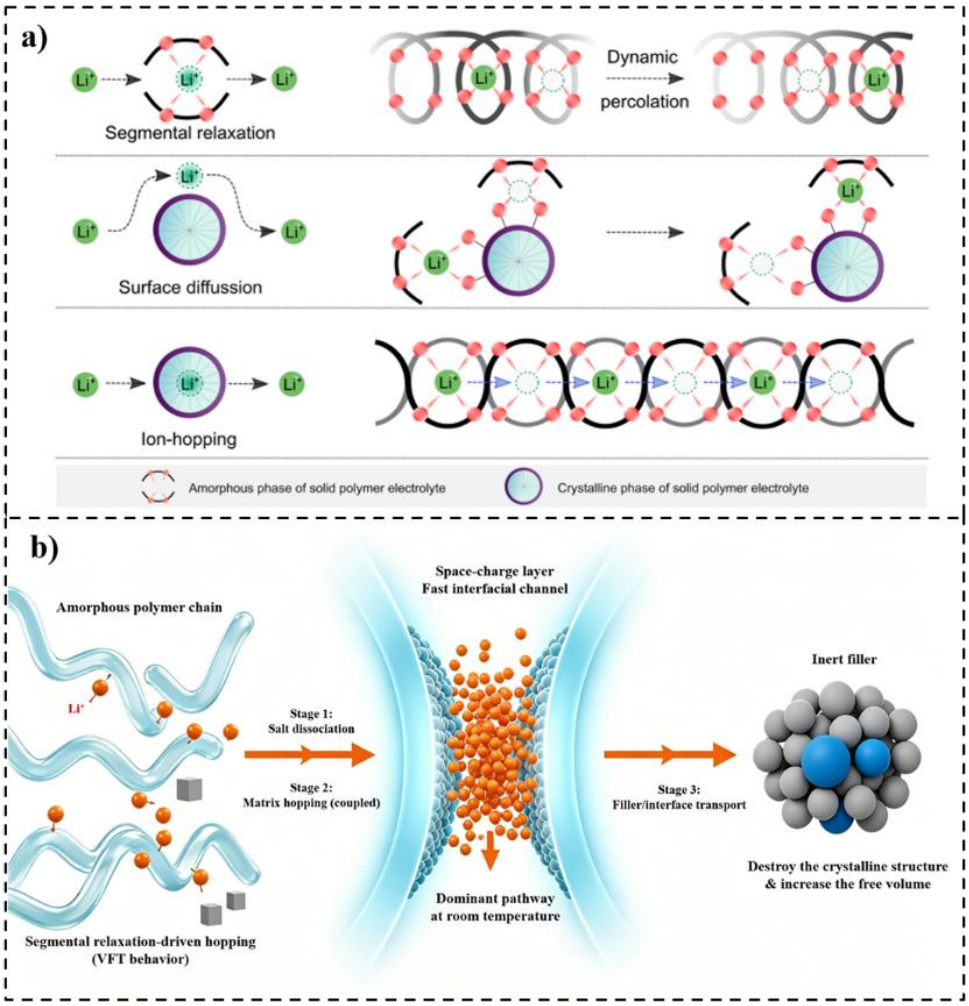

Fig 3. (a) Transport of ionic species in different coupled SPEs [11]. (b) Schematic illustration of the multi-pathway ion conduction mechanism in CSPEs (Picture credit: Original).

Li^+ transport in CSPEs follows a multi-step dynamic mechanism. It begins with dipole–dipole interactions between Li^+ and polar functional groups in the polymer backbone, which drive the dissociation of lithium salts into free Li^+ and anion. This element charge dissociation goes further along the polymer-filler interfaces by a local interfacial polarization due to neighbor inorganic additives. After dissociation, the liberated Li^+ temporary form as coordination bonds with the electron-donating sites in the amorphous regions in the polymer. The long-range migration underpins a continuous cycle of bond breaking and formation, which is closely associated with the local motions and conformational rearrangements of polymer segments. This temperature-dependent phenomenon follows the Vogel-Fulcher-Tammann (VFT) relation. Inorganic fillers play multi-dimensional auxiliaries according to their chemical nature. For example, the inclusion of structurally inert phase such as Al_2O_3 and SiO_2 are applied to disrupt the long range crystalline order of the polymer host to expand free volume, while alleviating the restrictions on segmental mobility. Active ion conductors such as LLZO and LATP achieve the continuous transport pathways across the grain boundary or particle surface. Moreover, the functional fillers configured by Lewis acid cation are different from two mechanisms, which chemically fix the incoming anions which enhanced the Li^+ transference number and mitigating the adverse concentration polarization during cycling [12,13].

The conduction efficiency is affected by polymer crystallinity, filler dispersion, salt dissociation, interfacial compatibility and temperature. Elevated temperature enhances segmental relaxation, reduces Tg and crystallinity, expands interconnected amorphous pathways and lowers Li^+ hopping energy barriers, boosting conductivity. The atomistic details of this network have been captured by DFT and MD simulations. These computational models show that polar interfaces can effectively weaken Li^+ anion interactions, thereby promoting salt dissociation within local transport channels. As a result, Li^+ transport in CSPEs proceeds through a hybrid, multipathway network that couples bulk polymer relaxation with filler-induced interfacial diffusion. However, current studies still struggle to resolve the real-time dynamics of these hybrid systems. In particular, the local charge distribution at the polymer–filler interface has not been quantitatively determined, and the evolution of percolation pathways during electrochemical cycling remains difficult to track. To establish a more accurate structure–property relationship, the field needs to move beyond static models toward operando characterization and multiscale coupling approaches [12–14].

4. Design and Modification Strategies of CSPEs

Rational design and targeted modification of CSPEs constitute the core strategy to address the performance bottlenecks of ASSBs, including low ionic conductivity (RT), unstable electrode-electrolyte interfaces, uncontrolled lithium dendrite propagation and insufficient mechanical robustness. Significant advances have been made in CSPEs modification based on advanced material design principles, interface engineering and structural regulation. This section systematically summarizes polymer matrix engineering, inorganic filler functionalization, lithium salt and additive optimization and structural innovation of CSPEs, combined with representative high-level publications to clarify the structure–activity relationship and performance enhancement mechanism, as illustrated in Figure 4.

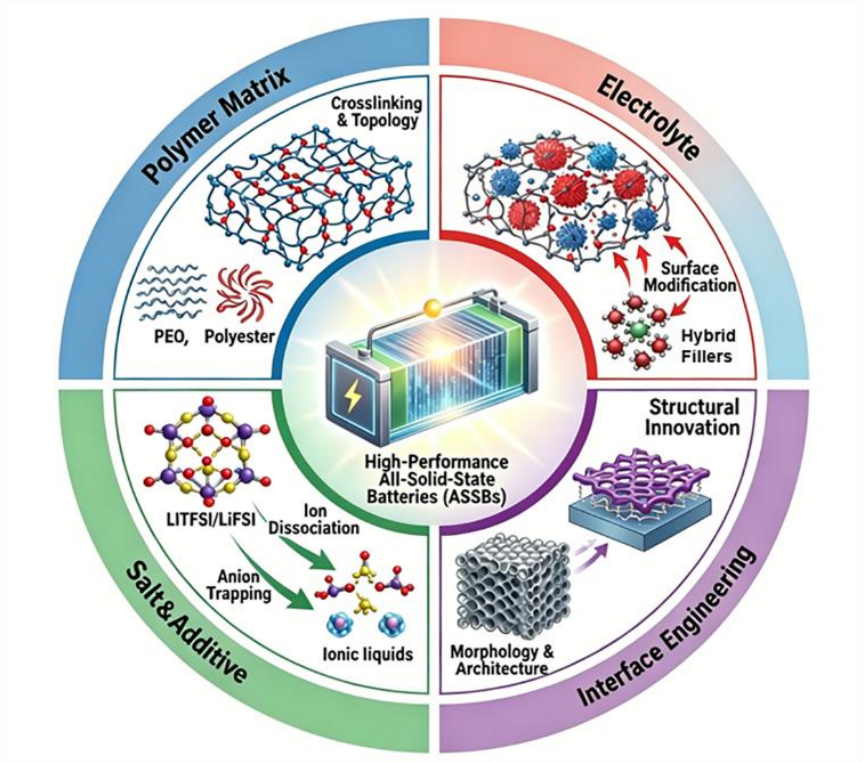

Fig 4. Multidimensional Design Strategies for CSPEs towards High-Performance ASSBs (Picture credit: Original).

4.1. Polymer Matrix Modification

Crosslinking modification is a core strategy to mitigate the inherent limitations of linear polymer electrolytes, including high crystallinity, low ionic conductivity (RT), poor thermal stability and insufficient mechanical strength. By constructing covalently bonded three-dimensional networks, crosslinking disrupts the ordered packing of polymer chains, reduce crystallinity and increases amorphous domain content. This structural regulation simultaneously enhances ionic conductivity at ambient temperature, high-temperature dimensional stability and mechanical modulus, enabling efficient Li^+ transport and stable electrode-electrolyte interfaces. On this basis, targeted engineering of polymer matrices, mainly including polyether, polyester, polysiloxane and poly (ionic liquid) (PIL) systems, has yielded a series of milestone breakthroughs. For polyether-based CSPEs, Sun et al. realized double-salt crosslinking of PEO via methylene radical reaction, achieving greatly widened electrochemical window (4.8 V) and high ionic conductivity up to 0.57 mS cm^{-1} at 30 °C. Recent studies on CSPEs show that polymer chemistry, network topology, and functional design can be adjusted together to improve electrolyte performance [15]. In ester-based systems, Fang et al. prepared in situ cross-linked, ester-rich electrolytes in which AlF3-assisted anion immobilization enabled a high Li^+ transference number of 0.67 and favorable compatibility with Li metal [16]. A similar strategy was explored in polysiloxane-based CSPEs, who constructed comb-like cross-linked networks bearing pendent PEO chains. The internal plasticization provided by these side chains increased the ionic conductivity by nearly one order of magnitude. For PIL-based electrolytes, Lin et al. incorporated zwitterionic liquid segments into cross-linked networks, obtaining a Li^+ transference number of 0.78 together with stable interfacial behavior toward Li metal [17].

Beyond the choice of polymer backbone, regulation of network topology has also played an important role. AB cross-linked polymers (ABCPs), semi-interpenetrating networks (semi-IPNs), and interpenetrating networks (IPNs) have been used to overcome the limitations of simple cross-linked networks. Guo et al. further developed double-network IPN CSPEs with a self-plasticizing effect, which improved thermal stability and resistance to Li dendrite growth. Functional modification provides another route to broaden the application scope of CSPEs [18].

4.2. Inorganic Filler Modification

Inorganic filler functionalization was the most widely exploited strategy for the optimizing of the overall performance of CSPEs. Inert ceramic fillers, active Li^+-conducting fillers and emerging two-dimensional functional fillers can effectively suppress polymer crystallization, construct additional ion-transport pathways, ameliorate electrode–electrolyte interfacial contact and reinforce mechanical strength. Meanwhile, directional arrangement of fillers has been proven to optimize ion-transport efficiency, and the synergistic effect of hybrid fillers integrates crystallinity reduction, ion conduction and interface stabilization, thus breaking the performance limit of single-filler CSPEs. Benefiting from the development of nanomaterial technology, Du et al. highlighted that novel functional fillers including MOFs, COFs and two-dimensional materials have been widely adopted, providing new opportunities for constructing high-stability CSPEs [19].

4.3. Lithium Salt Optimization and Additive Modification

Lithium salt optimization and additive modification play crucial roles in determining Li^+ dissociation extent, ion transport dynamics and interfacial formation behavior in CSPEs. Zhou et al. reported that LiTFSI is predominantly used in PVDF-HFP/LLZTO-based CSPEs and the in-situ formed $Li_xTaO_xF_{5-x}$ (LTOF) ion bridges via competitive coordination effects significantly reduce the LiTFSI dissociation energy barrier from 5.59 eV to 2.06 eV, increasing the fraction of free $TFSI^-$ to 60.1% without severe ion aggregation [20]. Feng et al. demonstrated that the combined utilization of LiTFSI and LiFSI in PEO-MOF-ionic liquid electrolytes offers complementary advantages, where LiTFSI ensures thermal stability and LiFSI promotes ionic conductivity [21]. Li et al. showed that the lithium salt concentration has a pronounced influence on the performance of gradient PDOL electrolytes [22]. An appropriate salt concentration helps maintain a balance between charge-carrier concentration and polymer segmental relaxation, whereas excessive salt loading can promote salt aggregation and lead to performance degradation. Functional additives can be used to further regulate the local electrolyte structure and interfacial chemistry. For instance, confining ionic liquids within amino-functionalized UiO-66-NH_2 nanoparticles drastically disrupts local polymer crystallinity to depress Tg, which simultaneously accelerates lithium salt dissociation and elevates ambient ionic conductivity. The bulk-phase Li^+ transport network is also supported by interfacial reactions at the Li-metal side. Decomposition of lithium salts generates in situ passivating species, leading to the formation of a compact SEI rich in LiF, Li_3N, and Li_2S. Such an interfacial layer can suppress parasitic reactions and help regulate local Li deposition, which is important for mitigating dendrite growth. Meanwhile, Lewis acidic Zr^{4+} sites in the MOF act as anion-trapping centers. Their interaction with anions restricts anion migration and increases the Li^+ transference number, contributing to lower polarization and better high-rate performance.

4.4. Structural Design of CSPEs

Structural regulation has become an effective approach to optimize CSPEs. For instance, Feng et al. fabricated porous PAN/PMMA scaffolds by electrospinning [21]. This porous structure provides a high specific surface area and continuous ion-transport channels, facilitating Li^+ diffusion and improving electrode–electrolyte interfacial contact.t. Li et al. focused on the mechanical stability of the interface and designed a multifunctional gradient CSPE with a solid-to-gel transition and an LLZTO modification layer [22]. By buffering local interfacial strain and lowering charge-transfer impedance, this structure maintains stable contact with the electrode even during mechanical deformation. Zhou et al. introduced amorphous LTOF ion-bridged structures onto LLZTO particles, forming three interconnected Li^+ transport pathways with reduced migration barriers [20]. The electrolyte achieved ionic conductivity of 1.21 mS cm^{-1}. Besides structural construction at the physical level, reversible chemical interactions, such as dynamic covalent bonds and supramolecular bonds, can also be used to develop self-healing CSPEs. These interactions help repair microcracks and reduce interfacial delamination, thereby improving the cycling durability and operational

reliability of ASSBs. These advanced structural design strategies, integrated with optimized lithium salt systems and functional additives, provide feasible solutions for the practical application of CSPEs.

5. Conclusion

CSPEs have become attractive core materials for high-safety and high-energy-density ASSBs, as they rationally integrate the excellent flexibility, processability and interfacial wettability of polymer matrices, thus overcoming the inherent defects of pure SPEs and ISEs. This review systematically summarizes the basic composition and ion conduction mechanism, comprehensively discusses the mainstream modification strategies including polymer matrix engineering, inorganic filler functionalization, lithium salt and additive optimization and structural innovation and emphatically analyzes the interfacial problems between CSPEs and electrodes as well as the corresponding regulation schemes. At present, the research of CSPEs and their comprehensive electrochemical properties has been significantly improved. Nevertheless, the large-scale practical deployment of CSPEs still encounters several critical bottlenecks.

At present, the ionic conductivity (RT) of most reported CSPEs is mainly in the range of 10^{-4}~10^{-5} S·cm^{-1}, while only a few systems achieve 10^{-3} S·cm^{-1}. It is generally inadequate for commercial application in terms of RT, the electrode-electrolyte interface is unstable with high interfacial impedance and continuous parasitic reactions, the inhibition effect on lithium dendrites needs to be further enhanced and the scalable preparation of defect-free thin films is restricted by high cost and poor batch consistency. Interdisciplinary efforts are needed to exploit new electrolyte materials and innovative structures, strengthen the interfacial interaction among different components and construct continuous ion transport pathways.

In addition, the in-depth understanding of ion transport dynamic mechanism and interfacial evolution law under operating conditions still needs to be strengthened by means of in-situ characterization and multi-scale theoretical simulation. In the future, breakthroughs should be focused on material innovation, precise interface engineering, low-cost scalable processing technology, high-resolution in-situ characterization and multi-scenario application expansion. Continuous optimization of material design and interface regulation will effectively solve the existing technical bottlenecks and strongly promote the industrialization process of CSPE-based ASSBs.

Compared with traditional liquid electrolytes, current CSPEs, especially those with complex architectures, suffer from high manufacturing costs. Future optimization should adopt raw materials free of rare and precious elements, simplify synthetic routes and design optimized composite structures to reduce raw material and energy consumption. Scalable production must be realized and technological compatibility between CSPEs and electrodes, as well as other internal components, must be guaranteed as well. Meanwhile, a reasonable balance between mechanical performance (including strength, thickness and elasticity) and practical energy density needs to be maintained. Furthermore, there is an urgent need to advance protective protocols for lithium metal storage and transportation and to establish consistent evaluation benchmarks for the performance, safety and recyclability of CSPEs and ASSBs, thereby promoting the transformation of laboratory research into practical commercial applications.

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