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Advancements in Gel Polymer Electrolytes: From Structural Engineering to Functional Applications

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Abstract

Gel polymer electrolytes (GPEs) have emerged as a promising class of quasi-solid-state electrolytes that combine the liquid-like interfacial compatibility of conventional liquid electrolytes with the mechanical integrity and leakage resistance of solid-state systems. Despite these advantages, their deployment in high-energy-density lithium batteries is still constrained by insufficient room-temperature ionic conductivity, low lithium-ion transference number (t_{Li+}), limited mechanical robustness, and unstable electrode/electrolyte interfaces. This review provides a mechanism-guided overview of recent advances in GPEs, with particular emphasis on the coupled regulation of ionic conductivity and t_{Li+}. First, the compositional features, ion-transport mechanisms, and key electrochemical evaluation metrics of GPEs are summarized to establish a unified structure-property framework. Subsequently, three major modification strategies are critically discussed: polymer-matrix engineering for crystallinity suppression and pathway construction, single-ion-conducting and anion-regulating designs for enhancing t_{Li+}, and functional filler incorporation for simultaneously improving mechanical strength, interfacial stability, and ion transport. Representative examples based on cross-linked networks, high-entropy gel systems, porous/aligned polymer frameworks, anion-anchored polymer electrolytes, boron-based anion receptors, inorganic nanofillers, metal-organic frameworks, and covalent organic frameworks are discussed in relation to their underlying mechanisms. Finally, the application prospects of GPEs in high-energy lithium batteries, flexible/wearable devices, and intelligent energy-storage systems are analyzed. Future research directions, including data-driven electrolyte design, scalable in situ manufacturing, and multifunctional integration, are proposed to guide the development of next-generation high-performance GPEs.

Keywords

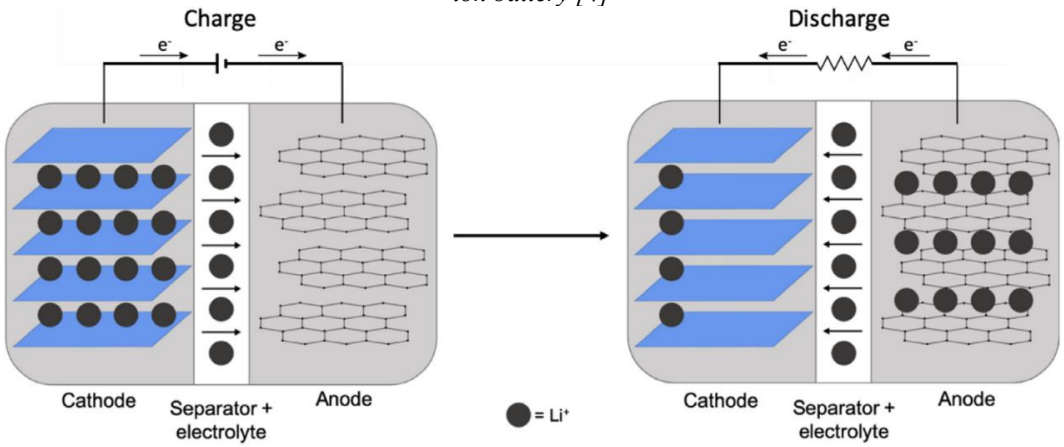
gel polymer electrolyte, lithium-ion battery, ionic conductivity, lithium-ion transference number, polymer matrix, functional filler, single-ion conductor

1. Introduction

The rapid expansion of electrified mobility, portable electronic devices, and grid-scale energy storage has fueled a pressing need for rechargeable batteries that offer high energy density, extended cycle lifespan, and dependable safety simultaneously. Lithium-ion batteries (LIBs) have been recognized as one of the most

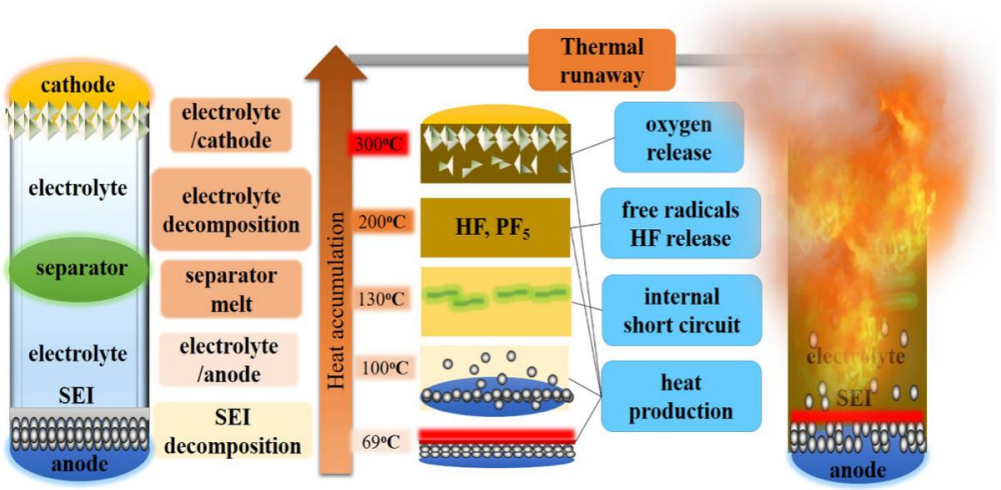
effective electrochemical energy-storage technologies because of their high operating voltage, mature manufacturing base, and wide applicability across consumer devices, electric automobiles, and intelligent power grid [1-3]. In a typical LIB, lithium ions shuttle reversibly between the cathode and anode through the electrolyte, while electrons travel through the external circuit. The electrolyte is therefore not merely an ion-conducting medium; it determines interfacial charge-transfer kinetics, solid-electrolyte interphase (SEI) formation, thermal stability, and long-term cycling behavior [4, 5].

Figure 1: Schematic illustration of lithium-ion and electron transport during charge/discharge processes in a lithium-ion battery [4]



Conventional liquid electrolytes (LEs), usually composed of carbonate solvents, lithium salts, and performance-enhancing additives, provide high ionic conductivity and excellent electrode wetting [6-9]. However, their volatility, leakage risk, flammability, and involvement in thermal runaway remain serious safety concerns, particularly under abuse conditions such as overcharging, mechanical damage, elevated temperature, or internal short-circuiting [7, 10-12]. Solid-state electrolytes (SSEs) have been intensively explored to overcome these safety issues because they offer improved thermal robustness, flame retardancy, and mechanical integrity [1, 13, 14]. Nevertheless, rigid solid-solid interfaces often plagued by high interfacial impedance, weak electrode/electrolyte adhesion, and interfacial debonding during cycling, which restrict ion transport and reduce practical cell performance [15-18].

Figure 2: Progressive thermal-runaway processes and electrolyte-related safety hazards in lithium-ion batteries [7]



Gel polymer electrolytes (GPEs) provide an intermediate and highly attractive solution. A typical GPE comprises a three-dimensional cross-linked polymer network that hosts lithium salt dissolved in an organic solvent and, when needed, additives or fillers [18-20]. Such a quasi-solid-state structure allows GPEs to retain the high ionic conductivity and interfacial wetting of LEs while improving leakage resistance, mechanical stability, and safety. Since Feuillade and co-workers first reported ion-conductive macromolecular gels for

lithium systems, [21] GPEs have evolved from simple solvent-containing polymer gels to multifunctional ion-transport platforms designed through polymer chemistry, interface engineering, and nanostructured filler incorporation.

Despite their promise, GPEs still face several key bottlenecks that hinder their application in high-energy-density lithium batteries. First, many polymer matrices exhibit partial crystallinity and restricted segmental motion at room temperature, resulting in inadequate ionic conductivity. Second, because both Li+ and counter anions can migrate in conventional dual-ion systems, the lithium-ion transference number (tLi+) is typically low. A low tLi+ leads to concentration polarization, uneven lithium deposition, and dendrite formation, thereby accelerating capacity decay and safety failure [22, 23]. Third, swollen gel networks may lack sufficient mechanical strength, while the electrode/electrolyte interface may degrade through interfacial side reactions, resistive SEI formation, and volume-change-induced contact loss.

Previous reviews have summarized the preparation methods, constituent materials, and general applications of GPEs [18, 20, 22, 24, 25]. However, for high-level SCI publication, a review should not simply list materials and examples; it should identify a clear scientific question and organize the literature around a mechanism-oriented framework. Accordingly, this review focuses on the central question: how can the structure of GPEs be rationally regulated to achieve the simultaneous improvement of ionic conductivity, tLi+, interfacial stability, and mechanical robustness. To address this question, we first summarize the composition and evaluation metrics of GPEs, and then discuss three major design strategies: polymer-matrix engineering, single-ion/anion-regulating modification, and functional filler incorporation. Finally, we analyze emerging applications and outline future directions in artificial-intelligence-assisted design, scalable manufacturing, and multifunctional integration.

2. Fundamentals of Gel Polymer Electrolytes and Vital Evaluation Metrics

2.1 Electrolyte Categories and the Role of GPEs

Lithium batteries electrolytes systems can generally be classified into liquid, solid, and gel-state systems according to their composition, physical state, and ion-transport pathway [6, 26, 27]. Liquid electrolytes are widely used in commercial LIBs because they provide rapid Li+ transport, low viscosity, and effective electrode wetting. They are commonly formulated from carbonate or ether-based organic solvents, lithium salts such as LiPF6 and LITFSI, and functional additives that regulate SEI chemistry and electrochemical stability [7-9]. However, their flammability and leakage risk motivate the development of safer alternatives.

SSEs, including inorganic ceramics, sulfides, oxides, and solid polymer electrolytes, can improve safety by replacing volatile liquid solvents [13, 14]. Nevertheless, the practical use of SSEs is limited by high solid-solid interfacial resistance, fabrication complexity, brittleness, and incompatibility with conventional roll-to-roll battery manufacturing [15-17]. In comparison, GPEs bridge the gap between LEs and SSEs. The liquid phase ensures efficient ion conduction and wetting, whereas the polymer framework suppresses leakage, maintains dimensional stability, and provides a platform for chemical and structural modification [19, 20].

2.2 Composition and Ion-transport Mechanism of GPEs

A GPE usually contains four functional components: a polymer matrix, a lithium salt, a liquid plasticizer or solvent, and optional fillers or additives. The polymer matrix acts as a structural scaffold and ion-coordination environment. Common matrices include poly(ethylene oxide) (PEO), polyacrylonitrile (PAN), poly(vinylidene fluoride) (PVDF), poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP), poly(methyl methacrylate) (PMMA), poly(vinyl chloride) (PVC), and poly(vinyl ethylene carbonate) (PVEC) [28-34]. These polymers contain polar functional groups such as ether oxygen, cyano, carbonyl, and fluorinated groups that interact with lithium salts and influence ion dissociation, segmental dynamics, and electrolyte uptake.

Ion transport in polymer-containing electrolytes is strongly coupled with polymer-chain mobility. In semi-crystalline matrices, Li+ migration mainly occurs in amorphous regions where segmental motion can assist ion hopping and solvation-shell rearrangement [35-37]. Crystalline domains reduce free volume and restrict chain dynamics, thereby increasing ion-transport resistance. Therefore, suppressing crystallinity, lowering

glass-transition temperature, increasing amorphous content, constructing liquid-rich pathways, and introducing directional transport channels are key routes to enhancing ionic conductivity. In addition, the motion of anions must be regulated because excessive anion mobility causes concentration polarization and reduces the effective Li+ flux across the cell [23, 38].

2.3 Critical Performance Indicators

For practical lithium batteries, GPEs must satisfy multiple requirements rather than a single conductivity target. The first key parameter is room-temperature ionic conductivity, commonly obtained by using an electrochemical impedance spectroscopy method on a cell assembled with stainless steel blocking electrodes. The conductivity could be calculated by:

$$\sigma=\frac{L}{R_b\times S} \tag{1}$$

The second key parameter is the lithium-ion transference number, which indicates the fraction of total ionic current carried by Li+. This parameter is commonly evaluated via potentiostatic polarization experiments conducted in symmetric lithium-metal cells and calculated according to the Bruce-Vincent-Evans equation:

$$t_{Li^+}=\frac{I_{ss}(\Delta V-I_0R_0)}{I_0(\Delta V-I_{ss}R_{ss})} \tag{2}$$

In addition to ionic conductivity and t_{Li+}, electrochemical stability window, thermal stability, mechanical strength, electrolyte uptake, interfacial impedance, and cycling stability must be considered. A high-performance GPE should provide sufficient ion conduction at room temperature, suppress anion-dominated current, maintain stable contact with both cathode and anode, resist oxidative and reductive decomposition, and tolerate mechanical deformation or electrode volume changes during cycling.

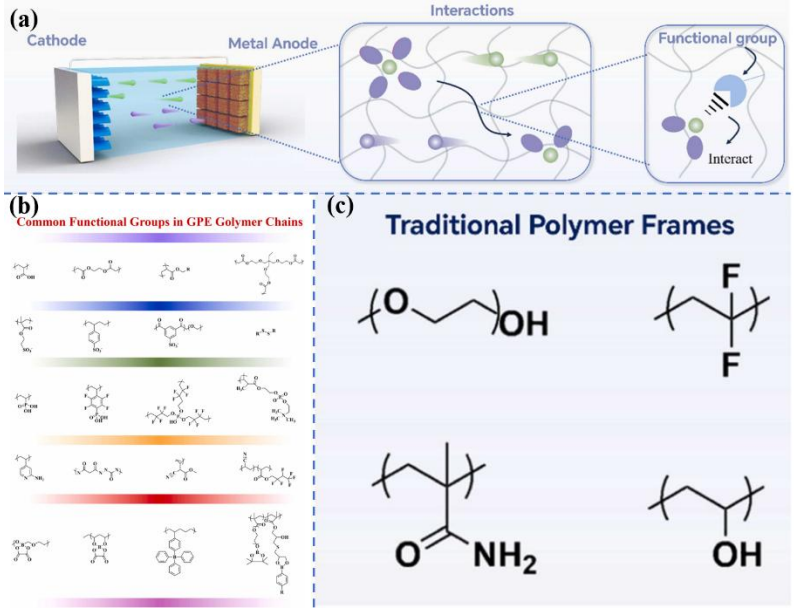
Table 1: Main limitations of GPEs and mechanism-oriented modification targets

Limitation	Mechanistic origin	Impact on batteries	Modification target
Low ionic conductivity	Restricted segmental motion, high crystallinity, insufficient salt dissociation, tortuous transport pathway	Poor rate capability and reduced power output	Suppress crystallinity, increase amorphous content, build liquid-rich or aligned ion channels
Low t_{Li+}	Fast anion migration and weak anion-polymer interaction	Concentration polarization, uneven Li deposition, dendrite growth	Anchor anions, introduce anion receptors, construct single-ion conducting networks
Mechanical fragility	Excessive solvent uptake and weak swollen polymer networks	Electrolyte leakage, deformation, short circuit and unstable cycling	Cross-linking, reinforcement, dynamic bonding, mechanically robust fillers
Interfacial instability	Side reactions, unstable SEI, poor electrode/electrolyte contact and volume-change-induced delamination	High impedance, capacity fading, shortened service life	In situ polymerization, interfacial chemistry regulation, adaptive/self-healing gel networks

3. Polymer-matrix Engineering for High Ionic Conductivity

The polymer matrix determines the basic mechanical framework, solvation environment, and segmental dynamics of GPEs. Functional groups such as ether oxygen, carbonyl, cyano, sulfonate, phosphate, amino, boron-containing groups, and disulfide bonds can interact with Li+ or counter anions and thereby regulate salt dissociation, ion mobility, and interfacial compatibility [39]. However, many conventional polymer matrices are semi-crystalline and structurally fixed, making their intrinsic ion transport insufficient for demanding high-rate batteries. Therefore, polymer-matrix engineering should focus on two interconnected goals: reducing crystallinity to activate segmental-assisted ion migration and constructing efficient ion-transport pathways.

Figure 3: Functional groups and polymer frameworks used in GPEs. (a) Interaction between ions and functional groups. (b) Representative functional groups in GPE polymer chains. (c) Common polymer matrices for GPEs [39]



3.1 Suppressing Crystallinity Through Cross-linked and High-entropy Networks

Ion transport in GPEs occurs primarily in amorphous regions, whereas crystalline domains restrict chain motion and impede Li+ migration [35-37, 40]. Cross-linking is a widely used strategy to disrupt regular chain packing and suppress crystallization. Cross-linked networks introduce junction points, structural defects, and spatial constraints, all of which hinder the formation of ordered crystalline regions and increase amorphous free volume [41, 42]. For example, PMMA-based chemically cross-linked gel networks fabricated via in-situ polymerization with controlled MMA/NPGDA ratios produced homogeneous amorphous ion-conducting phases. At an optimized MMA:NPGDA ratio, the resulting GPE achieved an ionic conductivity of 1.39 mS·cm-1 and an elevated tLi+ of 0.705 at room temperature [43].

Cross-linked networks can also enhance performance beyond crystallinity suppression. Functional groups within the network may interact with anions through electrostatic interactions, hydrogen bonding, steric hindrance, or Lewis acid-base coordination interactions, thereby reducing anion mobility and promoting lithium-salt dissociation [44, 45]. Dynamic cross-linked systems, such as polyrotaxane-based ionogels, further introduce reversible bonding and mechanical adaptability, enabling improved ion conduction and self-adjustable mechanical stability [46].

High-entropy design provides a newer route for stabilizing amorphous and disordered electrolyte structures. By introducing multiple components, high-entropy GPEs increase configurational entropy and disrupt long-range chain ordering [47-53]. Because ion transport is favored in amorphous domains, entropy-stabilized gel networks can improve ionic conductivity across a broad temperature range. For instance, high-entropy GPEs containing an optimized amount of dioxolane precursor exhibited ionic conductivity up to 4.42 mS·cm-1 at 25 °C [49]. Similarly, incorporating PAN into PVDF-HFP-based GPEs increases configurational disorder and introduces abundant nitrile groups, which strengthen dipole interactions, facilitate Li-salt dissociation, and enhance tLi+ [54].

3.2 Constructing Porous and Aligned Ion-transport Pathways

Although reducing crystallinity improves local segmental mobility, long-range ion transport also depends on pathway continuity and tortuosity. Pore-making strategies convert dense polymer matrices into sponge-like networks with interconnected micro- or nanopores. These pores can retain liquid plasticizers through capillary forces, forming liquid-rich channels that reduce activation energy and accelerate Li+ migration [55]. Modified SiO2-based biomimetic ant-nest ionogels provide a representative example: their hierarchical porous structure promotes smooth lithium-ion transport and forms a particle-enriched interface that helps suppress lithium

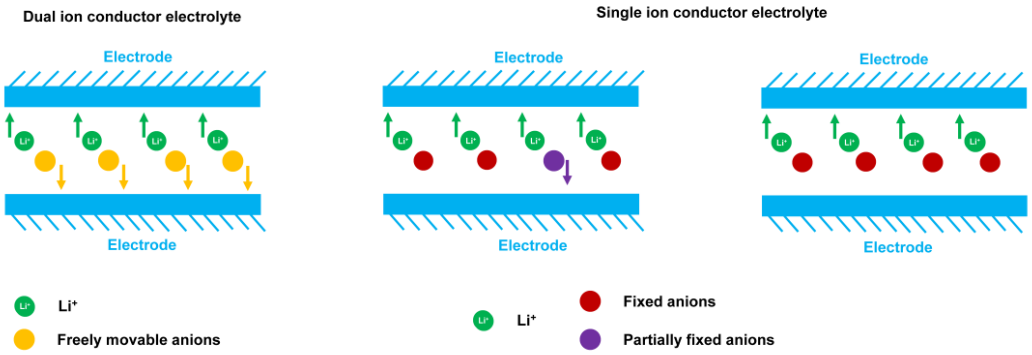
dendrite growth [56]. Nonsolvent-induced phase separation and electrospinning have also been widely used to construct porous polymer backbones for fast ion transport and enhanced interfacial stability [55, 57].

However, purely porous networks can be structurally disordered. Random pores may increase tortuosity, generate nonuniform ion flux, and create local concentration gradients. Directional alignment or stereospecific polymerization has therefore been developed to reduce transport tortuosity. In these designs, external fields, oriented scaffolds, or confined templates are used to align polymer chains, ceramic particles, or pore channels before or during gel formation [58, 59]. The resulting vertical or oriented pathways shorten Li+ migration distance and provide more direct conduction routes perpendicular to the electrode surface. For example, previous research has shown that infusing the GPE into vertically aligned carbon nanofibers functionalized with a silicon coating generates a highly permeable, low-tortuosity gel network, achieving an ionic conductivity of 2.2 mS·cm-1 at ambient temperature [60].

4. Lithium-ion Transference Number Regulation and Single-ion Conducting Strategies

A high ionic conductivity alone does not guarantee efficient battery operation. In conventional dual-ion electrolytes, both Li+ and anions migrate under an electric field, and anions may contribute a large fraction of the measured conductivity. Because only Li+ participates directly in lithium intercalation, plating, and stripping reactions, excessive anion migration causes concentration polarization, uneven Li+ flux, and dendritic deposition [23, 61-64]. Therefore, improving tLi+ is essential for lithium batteries with high-power capability and extended cycle stability. The core concept is to restrict anion mobility while maintaining or enhancing Li+ dissociation and migration.

Figure 4: Comparative analysis of electrolytes enabling dual-ion versus single-ion transport [65]



4.1 Anion Anchoring and Single-ion Conducting Polymer Networks

Single-ion conducting strategies aim to make Li+ the dominant mobile charge carrier by immobilizing anions on polymer backbones or inorganic scaffolds [38, 65-67]. In polymer-based single-ion conducting GPEs, anionic groups are covalently attached to the polymer backbones, leaving Li+ as the primary mobile species. The fixation strength of anionic groups can be regulated through molecular design, pKa tuning, and the selection of anion-coordinating functional units [44, 68-70]. Such designs can significantly suppress concentration polarization and improve lithium deposition uniformity.

A representative example is a lithium-based boron-centered fluorinated single-ion conducting GPE, in which anions are immobilized through molecularly designed covalent bonding [71]. Potentiostatic polarization combined with electrochemical impedance spectroscopy demonstrated a high tLi+ of 0.93, while temperature-dependent conductivity measurements indicated an activation energy value of 11.09 kJ·mol-1. The electrolyte also exhibited a extended electrochemical stability range, underscoring its potential in high-voltage lithium systems [71, 72].

Anions can also be immobilized by covalently grafting anionic groups onto inorganic nanoparticles. Because inorganic particles are essentially immobile within the gel matrix, anion-grafted fillers can lock anions while simultaneously expanding the amorphous phase and enhancing segmental mobility [73, 74]. For example, grafting sulfonated styrene units onto SiO2 nanoparticles improved room-temperature ionic conductivity to

2.28 mS·cm-1 and broadened the electrochemical stability window to over 4.8 V [75]. This strategy demonstrates the advantage of combining single-ion conduction with filler-induced polymer-matrix regulation.

Figure 5: Electrochemical properties of a representative single-ion conducting GPE: temperature-dependent ionic conductivity, electrochemical stability window, polarization current response, and impedance evolution [71]

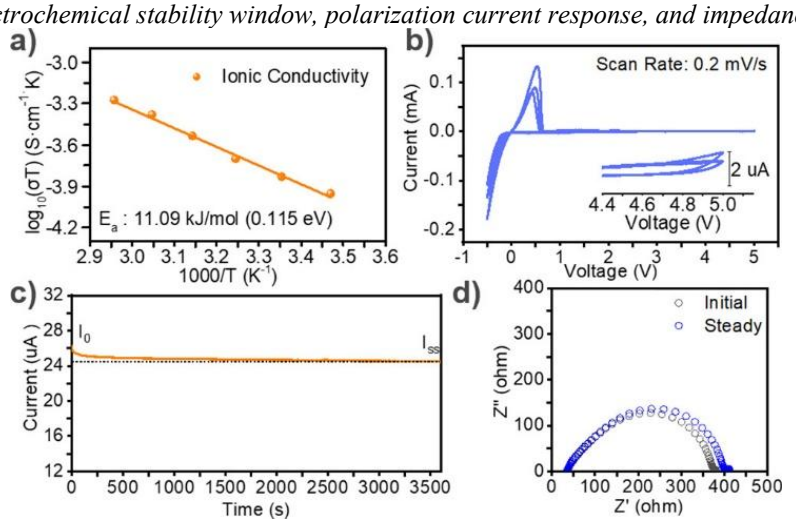


Table 2: Comparison of activation energies for representative polymer gel electrolytes

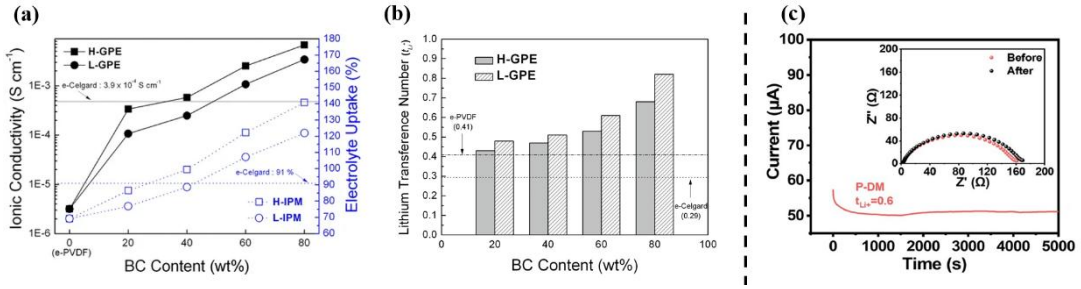
Polymer Gel Electrolyte	Plasticizer	Activation Energy (kJ/mol)
PPAB/PVDF-HFP[4]	PC/EC	9.86
LiPSI/PVDF-HFP[5]	EC/DMC	12.63
PAE-LiPFS[6]	DEC/EC/PC	17.9
PTHF700DA20[7]	EC/DEC	17.5
Poly(ETPTA)-$LiCF_3SO_3$[8]	TEGDME	16.1
LiBFSIE (this work)	EC/PC/FEC	11.09

4.2 Anion Receptors and Cation-anion Synergistic Regulation

Instead of covalently fixing anions, anion receptors can be introduced into GPEs to capture mobile anions through Lewis acid-base coordination, hydrogen bonding, dipole interactions, or supramolecular host-guest interactions [76-81]. Boron-containing compounds are among the most common Lewis-acidic receptors because the electron-deficient boron center can interact with Lewis-basic anions, weaken Li+-anion association, promote salt dissociation, and reduce long-range anion mobility [78-80].

Boron-containing cross-linkers in PVDF-based GPEs provide a representative example. The introduction of boron-based anion-trapping sites and ethylene oxide segments simultaneously enhances electrolyte uptake, ion dissociation, and anion immobilization, resulting in ionic conductivity up to 4.2 mS·cm-1 and tLi+ values as high as 0.82 at 30 °C [77]. Similarly, polar side chains containing amino or urea groups can form dense hydrogen-bonding networks with anions. Amine-functionalized dynamic covalent adaptive networks prepared by in situ cross-linking exhibit abundant anion-interacting sites and can increase tLi+ while maintaining interfacial compatibility [82].

Figure 6: Representative anion-regulating GPEs: ionic conductivity, electrolyte uptake, lithium-ion transference number, and polarization/impedance behavior [77, 82]



5. Functional Fillers for Multidimensional Performance Enhancement

Functional fillers are widely used to overcome the single-function limitation of polymer matrices. Appropriate fillers can reinforce mechanical strength, suppress polymer crystallinity, increase free volume, promote lithium-salt dissociation, immobilize anions, stabilize electrode/electrolyte interfaces, and create ordered ion-transport channels [83-85]. The effects of fillers depend strongly on their size, surface chemistry, Lewis acidity/basicity, porosity, and compatibility with the polymer matrix. Therefore, filler design should be discussed not only in terms of material category but also according to the specific function it provides.

5.1 Inorganic Nanofillers

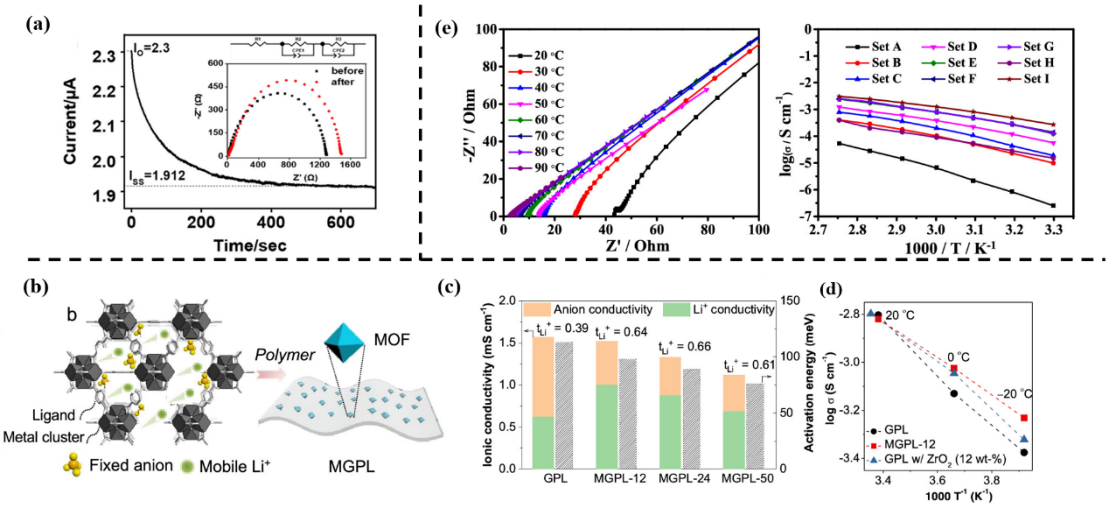
Inorganic nanofillers such as SiO2, Al2O3, TiO2, ZrO2, and graphene oxide (GO) can disrupt polymer-chain packing through steric hindrance and surface interactions, thereby reducing crystallinity and enhancing segmental mobility [83, 84, 86]. Surface Lewis-acidic sites can interact with anions or polymer functional groups, improving salt dissociation and increasing tLi+. In addition, rigid particles can reinforce swollen gel networks and improve dimensional stability. GO, for example, contains abundant oxygen-containing groups that interact with PVDF-HFP-based matrices and lithium salts. The incorporation of GO into a PVDF-HFP/LiTFSI/ionic liquid system increased tLi+ to 0.79 and improved overall electrochemical performance [83].

5.2 Porous Functional Fillers: MOFs and COFs

Porous crystalline frameworks, particularly metal-organic frameworks (MOFs) and covalent organic frameworks (COFs), have garnered growing attention owing to their synergistic integration of large specific surface area, customizable pore-surface chemistry, and well-defined ion-conduction pathway [84, 85, 87]. MOFs can offer accessible metal centers that function as Lewis acid sites to immobilize anions and facilitate lithium-ion migration. Their regular micropores reduce transport barriers and provide continuous pathways through the gel network. For example, Zr-based UiO-66 MOFs introduced into GPEs enhanced tLi+ from 0.39 to 0.66 while maintaining ionic conductivity above 10-3 S·cm-1 and reducing activation energy [85].

COFs are fully organic crystalline porous materials linked by covalent bonds [87]. Compared with many MOFs, they offer metal-free frameworks, low density, structural adjustability, and outstanding chemical resilience. Functional groups such as carboxyl, amino, sulfonate, or ether units can be incorporated into COF skeletons to regulate ion-solvent interactions and create specific lithium-ion pathways [4, 87]. COF nanochannels can also disrupt Coulombic ordering in ionic liquid-based gels and accelerate ion diffusion. For instance, TPB-DMTP-COF incorporated into a polymeric ionic liquid matrix produced a conductive composite ionic GPE with an ionic conductivity of 1.26 mS·cm-1 [86].

Figure 7: Representative filler-modified GPEs based on GO, MOF, and COF strategies, showing lithium-ion transference behavior, ion-channel regulation, conductivity, activation energy, and thermal/electrochemical properties [83-87]



6. Applications of Advanced GPEs

6.1 High-energy and High-safety Lithium Batteries

The rational design of GPEs enhances both safety and operational stability in high-energy lithium batteries. In high-capacity anode systems, such as silicon or lithium metal anodes, GPEs can form intimate electrode/electrolyte contact and accommodate volume changes through in situ polymerization or adaptive gel networks [88]. On the cathode side, high-voltage stability is essential for pairing GPEs with Ni-rich layered oxides or other high-voltage cathode materials. Dual-salt PVEC-based GPEs, for example, broaden the electrochemical stability window and suppress electrolyte decomposition under high-voltage operation [34]. Thermally responsive GPEs can further provide self-protection functions by regulating ion transport or phase behavior at elevated temperatures, thereby reducing thermal runaway risks [89, 90].

6.2 Flexible, Wearable, and Self-healing Devices

Flexible and wearable electronics require electrolytes that tolerate bending, stretching, compression, and accidental damage while maintaining stable ion transport [24, 91]. GPEs are particularly suitable for such applications because polymer networks can be designed with elasticity, adhesion, self-healing behavior, and shape conformability. Self-healing ionogels and GPEs based on reversible hydrogen bonds, ionic interactions, dynamic covalent bonds, or host-guest interactions can repair internal or external damage and restore electrochemical performance [45, 92-94]. Such properties are beneficial for flexible batteries, yarn-based supercapacitors, implantable devices, and integrated smart energy-storage systems.

7. Conclusions and Perspectives

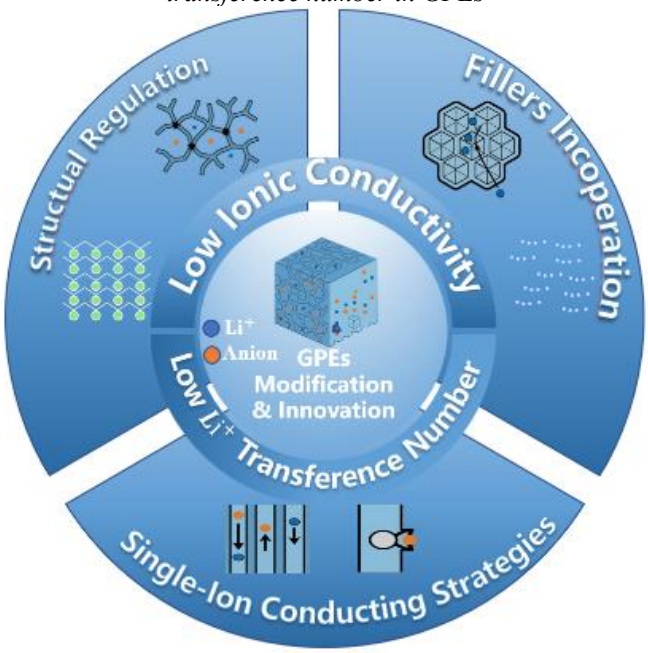
GPEs represent a highly promising electrolyte platform for next-generation lithium batteries because they integrate liquid-like ion transport and interfacial wetting with solid-like mechanical integrity and leakage resistance. This review has summarized recent advances in GPEs from the perspective of mechanism-guided structural design. Polymer-matrix engineering enhances ionic conductivity by suppressing crystallinity, increasing amorphous free volume, and constructing porous or aligned ion-transport pathways. Single-ion conducting and anion-regulating strategies improve t_{Li+} by immobilizing anions through covalent anchoring, Lewis acid-base interactions, hydrogen bonding, or receptor-mediated coordination. Functional fillers, including inorganic nanoparticles, MOFs, and COFs, provide multidimensional regulation of mechanical robustness, ion dissociation, anion mobility, and ordered transport channels. Together, these strategies demonstrate that high-performance GPEs require coordinated optimization of conductivity, t_{Li+}, mechanical stability, interfacial chemistry, and electrochemical compatibility.

Nevertheless, several challenges remain before advanced GPEs can be translated into large-scale high-energy batteries. First, many reported systems achieve high conductivity or high t_{Li+} under specific laboratory conditions, but their performance under realistic cell formats, high areal loading, lean electrolyte conditions, wide temperature ranges, and long-term cycling must be further validated. Second, the relationships among polymer chemistry, solvation structure, filler surface chemistry, interfacial reactions, and lithium deposition behavior remain insufficiently understood. Advanced characterization techniques, including in situ spectroscopy, operando electrochemical imaging, solid-state NMR, neutron scattering, and multiscale modeling, should be combined to reveal ion-transport and interfacial-evolution mechanisms. Third, many sophisticated GPE designs are difficult to scale because they rely on complex monomer synthesis, expensive fillers, multistep fabrication, or poorly controlled in situ polymerization.

Future research should therefore focus on three directions. The first is artificial-intelligence-assisted and data-driven electrolyte design. By constructing databases that include polymer matrices, monomers, solvents, salts, fillers, mechanical properties, transport parameters, and cell-level performance, machine learning can accelerate material screening and identify hidden structure-property relationships. The second is scalable and integrated manufacturing. Low-cost, continuous, and electrode-compatible processes, especially controllable in situ polymerization and roll-to-roll compatible gel formation, are essential for industrial implementation. The third is multifunctional expansion. Beyond conventional LIBs, GPEs can be designed for lithium-metal batteries, lithium-sulfur batteries, flexible/wearable electronics, biomedical power sources, self-healing devices, and batteries operating in extreme environments. Achieving these goals will require interdisciplinary

integration of polymer chemistry, electrochemistry, interfacial science, materials informatics, and battery engineering.

Figure 8: Graphical abstract. Mechanism-oriented strategies for improving ionic conductivity and lithium-ion transference number in GPEs



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Conflicts of Interest

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