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Gel polymer electrolytes: definitions, classification, rheology, and interfacial properties

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Lithium-based batteries have revolutionized the electrochemical energy storage field, where liquid electrolytes are most used. Despite having high energy density and ionic conductivity, unwanted Li dendrite formation, leakage, and safety issues are ongoing challenges. In addition, the continuous lithiation/delithiation during charging/discharging processes affects both electrodes and electrolytes in terms of crack formation in the electrode, solid electrolyte interphase (SEI), cyclic compression/elongation of the electrolyte, and detachment of the interface especially in the solid electrolyte, which all can be addressed by using appropriate gel polymer electrolytes (GPEs). Therefore, this perspective starts with the definition of gels from a rheological perspective, categorizing GPEs, and finally discussing rheological considerations in electrolyte design and interfacial phenomena at electrode–electrolyte junctions.

Introduction

Since the discovery of fire, energy has been the driving force behind development and evolution in societies. Also, finding a way to store energy has been of interest. For example, the first speculated electrochemical cell, called the Parthian galvanic cell dating back to ∼200 BC, was likely used for electroplating or as a primitive power source, highlighting early human interest in utilizing electrical energy.[1–4] In the 21st century, energy is powering everything from homes to industries. As demand grows, efficient energy storage devices become essential for ensuring a stable and sustainable energy supply. For instance, energy storage systems balance energy supply and demand, enable renewable energy integration, and power portable devices and electric vehicles.[5,6] Fig. 1 presents the chronological advancements in electrochemical storage devices from 200 BC to the present day.

Batteries, as the main energy storage devices, are composed of an anode, a cathode and an electrolyte, in which the

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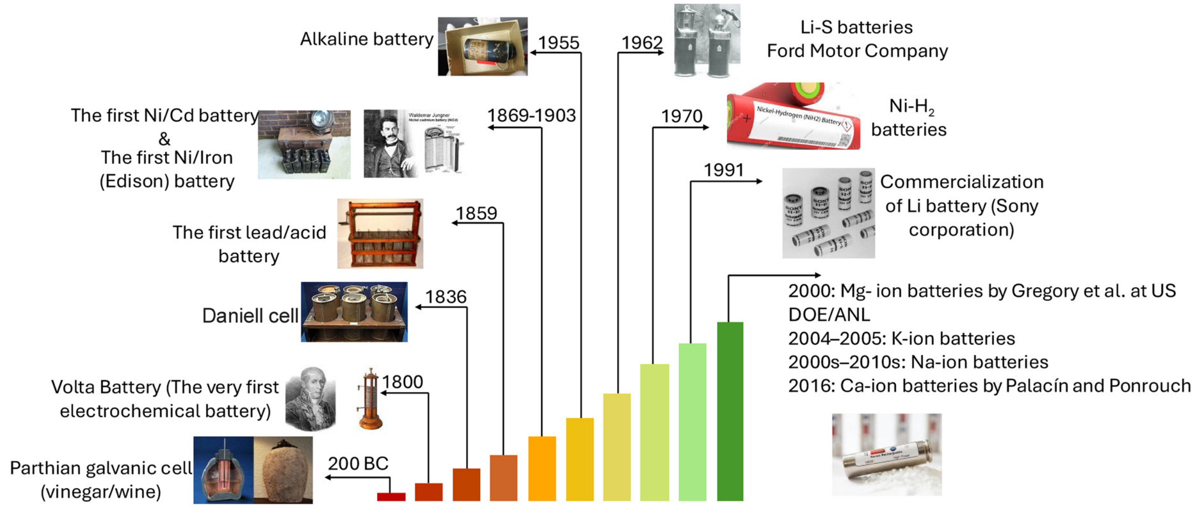

Fig. 1 History of electrochemical storage devices.

electrolyte plays a critical role alongside the electrodes by enabling charge carrier movements between the anode and cathode.[7,8] Liquid electrolytes have been the most common electrolytes used in batteries for decades due to their high ionic conductivities (10^{-3}–10^{-2} S cm^{-1}) and good contacts with electrodes (*i.e.*, good wetting).[9,10] Although the liquid electrolytes exhibit good electrochemical properties, they have some drawbacks, such as leakage, Li dendrite formation, poor mechanical properties, flammability, and toxicity.[10–16]

To address the above-mentioned challenges in liquid electrolytes, solid electrolytes were introduced into the electrochemical system. Solid electrolytes with a matrix of inorganic ceramics or polymers can provide some advantages such as high mechanical strength, simple packaging processes, and preventing dendrite formation as well as leakage.[17–19] The conductivity of a solid electrolyte (silver sulfide) was first observed by Faraday back in 1836, while β-alumina was the first commercially available solid electrolyte introduced in 1960.[20] Inorganic solid electrolytes, like oxides and sulfides, are known for their thermal stability and high ionic conductivity at elevated temperatures.[21,22] Beyond the inorganic solid electrolytes, solid polymer electrolytes (SPEs) gained attention due to their wide electrochemical window, non-brittleness, and good thermal stability. The first SPE was introduced in 1979.[20,23–26]

SPEs have been synthesized by using a polymeric host and a salt as the source of ions and in some cases mobile species for charge transport.[27] Despite having good electrochemical and mechanical properties, SPEs encounter issues such as low ionic conductivity and weak electrolyte–electrode interfaces.[28] Poly(ethylene oxide), PEO, is one of the most common polymers studied for electrolyte applications. However, PEO has a low ionic conductivity of 10^{-8} S cm^{-1} due to its semi-crystalline nature, resulting in poor battery performance.[10,29] High crystallinity in polymers hinders ion movements and reduces ion conduction.[10,26,29,30] The basic criteria for choosing a polymer host are high thermal and electrochemical stability, sufficient molecular weight to ensure good mechanical properties, low glass transition to accelerate the local chain movement and ion transport through the matrix, and functional groups that interact with ions, enhance salt dissolution inside the polymer and prevent dendrite formation.[31] However, all these features are not usually found at the same time in these polymers.

GPEs are developed to address low ionic conductivity and weak electrolyte–electrode interface issues in SPEs. Typically, to synthesize GPEs, a polymeric matrix is mixed with a liquid component (which will be discussed in the "Gel polymer electrolytes" section). While the polymer maintains strong mechanical integrity, the liquid component enhances diffusion of ions, providing high ionic conduction.[32,33] GPEs have low modulus and can be formulated to remain elastic under high deformations, which make them ideal candidates to be used instead of liquid electrolytes for addressing the leakage and dendrite formation issues.[34] While the lithiation/delithiation process has been extensively studied in the context of electrode behavior,[35–37] much less is known about its influence on electrolytes. In fact, this process is the primary source of deformation in electrolytes, which occurs repeatedly during battery charging and discharging throughout its lifetime. Therefore, this perspective highlights how lithiation/delithiation affects the electrolyte and electrolyte–electrode interface in the context of the overall stability and efficiency of batteries. We provide a deeper insight into how GPEs integrate mechanical resilience with electrochemical functionality by connecting the network structure and rheological response with interfacial stability.

The outline of this perspective is as follows. First, it is essential to restate a precise and consistent definition of gels from a physical viewpoint, because the term "gel polymer electrolytes" is often applied to systems that are not inherently gels. Many studies, for instance, have broadly classified liquid-supported membranes as GPEs.[38–40] However, such generalization does not always align with the physical definition of a gel. In other words, we consider GPEs as only those electrolytes that exhibit the fundamental features of gels, rather than all liquid-containing membranes. This distinction is important because it ensures consistency in terminology and avoids conflating materials with fundamentally different structural and functional behaviors. After revisiting the gel definition, we review the literature on GPEs that truly meet the gel criteria, providing a revised classification for comparing electrolyte design strategies across different studies.

Revisiting gel definitions

In the Merriam-Webster dictionary, a gel is defined as ''a colloid in a more solid form than a sol''. In a more scientific way, Jones defines gels as materials composed of connected subunits that form networks of macroscopic dimensions, in which bonding of subunits induces a liquid's (sol) transition to a gel with solid-like mechanical properties.[41] Based on the continuous structure of gels, Flory classified them into four categories: (i) gel mesophases (well-ordered systems); (ii) disordered covalent polymeric networks; (iii) disordered physically aggregated polymer networks with a local ordered structure; and (iv) particulate, disordered structures.[42] The main focus of this perspective is GPEs, thus, category (ii) and (iii) are mainly of interest.

Gels are prepared as two- or multi-component coherent systems, constituted by a solid (a gelator) and solvent(s), where the solid component creates a 3D network expanding throughout the liquid phase.[43,44] It is proposed that the solid network in gels traps and immobilizes the liquid phase of at least ~1 time and up to 1000 times of network mass, while having the ability to reversibly swell or deswell.[45] We believe the definition of a gel is independent of the amount of immobilized liquid and should be treated from a physical property viewpoint.

The gelation process is a thermodynamic phase separation (*e.g.*, temperature or solvent changes) that forms heterogeneous structures when full separation is prevented. The sol–gel transition point is crucial for characterizing the gelation process. The sol–gel transition can be induced by different stimuli, *e.g.*, temperature,[46] stress,[47] concentration or volume fraction,[48] time,[49] pH,[50] and light.[51] The most common ones are the temperature- and shear-induced gelation.

The first rheological criterion for identifying the gelation point was proposed by Winter and Chambon in the 1980s. They stated that, at the gel point, the storage modulus (G') and loss modulus (G'') should have the same scaling behavior with frequency, following the relationship $G'(\omega) \sim G''(\omega) \sim \omega^n$, in which n is a constant.[52,53] Therefore, tan(δ) becomes independent of frequency at the gelation point and $\delta = n\pi/2$ with $0 < n < 1$. If n reaches 1, the gel is viscous, while for n close to 0, the gel is elastic.[54] This behavior is schematically shown in Fig. 2A before, after and during the gelation. Before and after the gelation, $G'(\omega)$ and $G''(\omega)$ do not simultaneously correlate with ω^n.[55]

The sol–gel transition point may be shifted by the introduction of salts into the system. Depending on the ionic state of the polymer, the presence of salts can increase or decrease the gelation temperature. For charged polymers, the higher the salt concentration, the greater the gelation temperature.[57] Comparing the effect of ions on the gelation of polymers shows that the cation–polymer interactions play a critical role in triggering and modulating gelation. The gelation of κ-carrageenan offers a well-understood case of cation influence on the physical properties of polymer gels.[58] For example, divalent cations have a stronger effect on both gelation temperature and G' of the κ-carrageenan gel due to their ability to bind the carrageenan helices together.[59,60] For neutral polymer gels, increasing salt

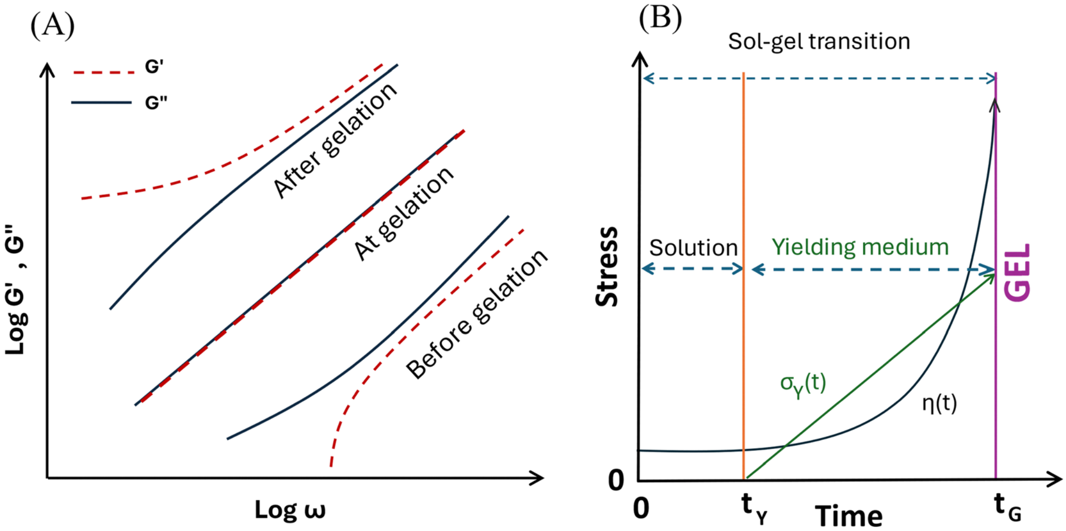

Fig. 2 (A) Schematic of viscoelastic properties before, after, and during gelation. During gelation, $G'(\omega)$ and $G''(\omega)$ scale with ω^n at all frequencies. Adapted with permission from H. H. Winter and M. Mours, *Neutron Spin Echo Spectroscopy Viscoelasticity Rheology*, 1997, 165–234. Copyright 1997 Elsevier. (B) A rheological model of gelation, representing the evolution in viscosity and stress during sol–gel transition.[56] Adapted with permission from A. Ya. Malkin and S. R. Derkach, *Curr. Opin. Colloid Interface Sci.*, 2024, **73**, 101844. Copyright 2024 Elsevier.

concentration deceases the gelation temperature, while leading to higher G'.[61–63]

Fig. 2B shows that the viscosity increases during gelation due to molecular interactions/reactions, leading to the formation of an elastoviscoplastic material characterized by a yield stress (σ_Y), which is the minimum stress needed to initiate flow. More comprehensively, σ_Y is defined as the material property that marks the shift from solid-like (recoverable deformation) to liquid-like (unrecoverable deformation) behavior.[64] Although theoretical criteria suggest that the gel is formed when a continuous network spans the entire system, in practice, the gel point is often determined by the loss of flow and/or appearance of yield stress.[65–67] More accurately, the material transitions from a fluid to an elastoviscoplastic state, existing in either gel-like (below σ_Y) or liquid-like (above σ_Y) behavior depending on the applied stress.[68,69] In this context, gel-to-sol transition can happen by increasing stress above yield stress, in which the 3D network can be reversibly or partially irreversibly disrupted by applying sufficient stress. The mechanically induced gel-to-sol transition is more probable in physical gels and chemical gels with very low crosslink density, while chemical gels with high crosslink density mechanically fracture before yielding. The linear viscoelastic properties of gels are well understood with their ideal behavior typically featuring a broad rubber-like plateau of $G'(\omega)$ accompanied by relatively low values of $G''(\omega)$, which confirms the semi-solid state of the gels.[70–72]

Gel polymer electrolytes

GPEs serve as both electrolytes and separators with flexibility, shape adjustability, and resilience for electrode volume changes.[73,74] GPEs support effective ion transport by providing interconnected ion pathways and enhance the performance, safety, and flexibility of advanced energy storage devices, which are visualized in Fig. 3.[75]

Some studies have referred to supported liquid membranes as GPEs.[38–40] However, these membranes do not always align with the fundamental definition of a gel. Maintaining a distinction is essential for consistent terminology and for preventing the misclassification of electrolytes with differing structural and functional properties. In the subsequent section, we briefly review the body of literature on polymer electrolytes that fulfill the gel criteria and organize them into categories based on their components, thereby providing an improved framework for comparing electrolyte design strategies across different studies.

Classification of GPEs

In GPEs, a polymeric scaffold is commonly employed as the primary matrix, providing substantial structural integrity and mechanical strength. To choose an effective polymer matrix, specific characteristics are required: (i) a high molecular weight (10^4–10^6 g mol^{-1}) for mechanical stability, (ii) a low glass transition temperature ($T_g < -30$ °C) enabling rapid segmental motion of polymer chains, (iii) functional groups or atoms that facilitate salt dissolution for improved ionic conductivity, (iv) a high thermal degradation threshold (>200 °C), and (v) a wide electrochemical stability range.[29] Therefore, the most commonly used polymers in GPEs are PEO,[31,76] poly(vinylidene fluoride) PVDF and poly(vinylidene fluoride-*co*-hexafluoropropylene) (PVDF-*co*-HFP),[77–79] poly(acrylonitrile) (PAN),[80] and poly(methyl methacrylate) (PMMA).[81,82] As illustrated in Fig. 4, GPEs are classified based on their fundamental components: the solvent or plasticizer system, the polymer host matrix, and the nature of the charge carriers.

Classification of GPEs based on solvent. Alongside the polymer host, the solvent plays a crucial role in GPEs. Selecting an appropriate solvent for dissolving the polymer and salt is essential as it remains trapped in the final gel. The solvent influences ionic conductivity and the stability as well as the

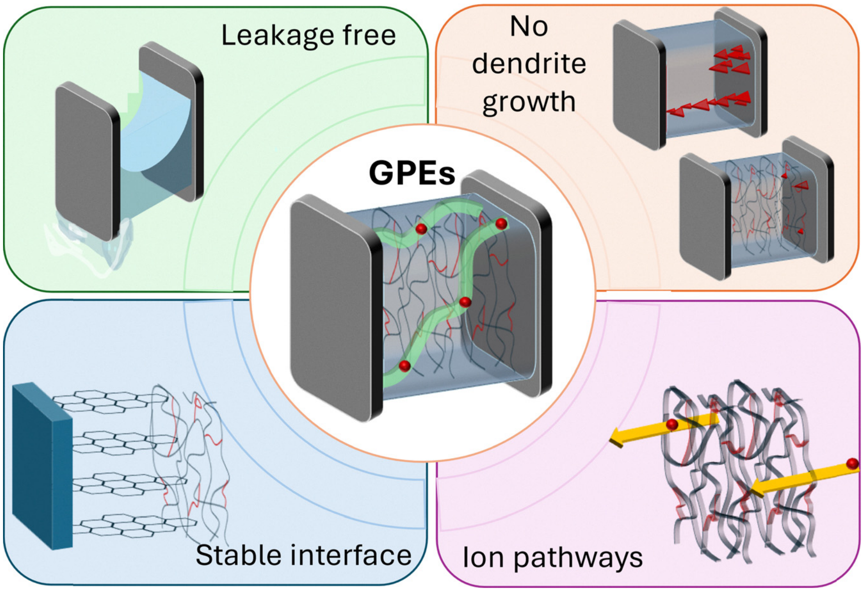

Fig. 3 Common properties of GPEs, which make them strong candidates for replacing conventional electrolytes.

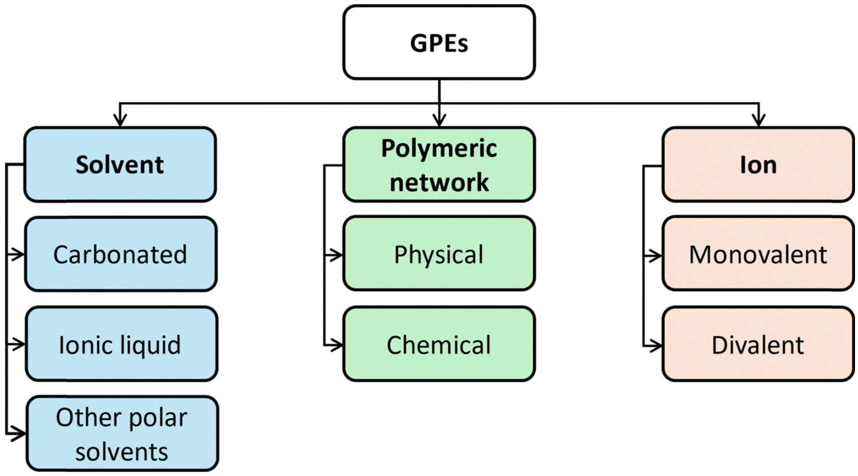

Fig. 4 GPE classification based on solvent, polymer network, and ion type.

mechanical and chemical properties of the GPEs.[83–85] Another important aspect is the ion–solvent interactions in GPEs that can hinder ion desolvation and slow down ion transport kinetics. This phenomenon also promotes solvent co-intercalation into electrode materials, which can degrade battery performance and stability.[86] Solvents are typically chosen among the cyclic carbonates,[6,87] glymes (*i.e.*, glycol ethers),[88,89] or ionic liquids (ILs).

Cyclic carbonates. An ideal solvent should have a high dielectric constant ($\varepsilon > 15$), a high donor number (>20) to promote ion dissociation,[90] and both chemical and electrochemical stability. The higher donor number enhances ion dissociation. Common organic solvents include EC, PC, DEC, DMC, EMC, dimethyl formamide (DMF), dimethyl sulfoxide (DMSO), and tetrahydrofuran (THF), for which the viscosity and dielectric constant are listed in Table 1. The donor number for cyclic carbonates is around 15 to 16, which is considered a low donor number, compared to the value of $\sim$30 for DMSO.[90,91]

Comparing dimethyl carbonate (DMC) with linear carbonates like ethyl methyl carbonate (EMC) and diethyl carbonate (DEC) in liquid lithium-ion electrolytes shows that despite similar viscosity and static permittivity, DMC exhibits 2–3 times higher ionic conductivity due to greater salt dissociation, enabled by polar *cis–trans* conformers. While viscosity impacts diffusivity, the higher ionic conductivity in DMC arises from more charge carriers due to improved salt dissociation even at low salt concentrations. This highlights that optimizing solvent conformational isomerism, alongside understanding viscosity and salt diffusion, is crucial for designing efficient battery electrolytes.[104] It should be noted that there are fluorinated solvents such as fluorinated ethylene carbonate (FEC)[105,106] and fluorinated dimethyl carbonate (FDMC) that offer high flame retardancy to reduce the flammability risks of carbonated solvents.[107] While these fluorinated compounds have gained interest as electrolytes, the growing concerns over the environmental impact of per- and polyfluoroalkyl substances (PFAS)[108] may eventually reduce their applicability at an industrial scale.

Table 1 Dielectric constant and viscosity of common liquid electrolytes and solvents in GPEs

Solvent	Dielectric constant	Viscosity (mPa s) (25 °C)
Ethylene carbonate (EC)	89.8[92]	1.9 (40 °C)
Propylene carbonate (PC)	66.1[93]	2.5[94]
Diethyl carbonate (DEC)	2.8[95]	0.749[96]
Dimethyl carbonate (DMC)	3.1[97]	0.59[98]
Fluorinated ethylene carbonate (FEC)	78.4[6]	4.4[99]
Ethyl methyl carbonate (FMC)	3.2[97]	0.65[98]
Dimethyl sulfoxide (DMSO)	47[100]	1.99[101]
Tetrahydrofuran (THF)	7.58[102]	0.46[103]

Ionic liquids. ILs offer significant advantages for GPEs due to their outstanding thermal and electrochemical stability, high ionic conductivity, non-volatility, non-flammability, and compatibility with various polymer matrices. IL-based GPEs are highly suitable for advanced energy storage and conversion applications.[109–111] Polymeric networks can immobilize ILs and form ionogels that are promising candidates for energy storage.[112] IL-based GPEs combine the beneficial properties of both aqueous and organic gelled electrolytes, including high ionic conductivity and a wide operating potential range of up to 3.5 V.[113–116]

Fig. 5 depicts the chemical structure of cationic and anionic parts of common ILs. ILs are widely studied in the literature due to their excellent electrochemical and thermal stability.[117–119] The thermal stability of ILs is influenced by both cation and anion types, typically decreasing in the order of imidazolium > pyridinium > pyrrolidinium > ammonium for cations and $[\mathrm{Tf_2N}]^- > [\mathrm{CF_3SO_3}]^- > [\mathrm{BF_4}]^- > [\mathrm{PF_6}]^- > [\mathrm{Br}]^-$ for anions.[120] Also, anion size significantly affects ion dissociation, meaning larger anions show weaker cation–anion coordination, which facilitates dissociation. Recent studies show that IL electrolytes containing bis(fluorosulfonyl imide) ($\mathrm{FSI^-}$) anions have higher ionic conductivity and lower viscosity than those with TFSI anions, attributed to the smaller size of $\mathrm{FSI^-}$.[118,121]

Classification of GPEs based on polymer networks

Physical networks. Typical physical networks are formed by non-covalent interactions, such as hydrogen bonding, ionic

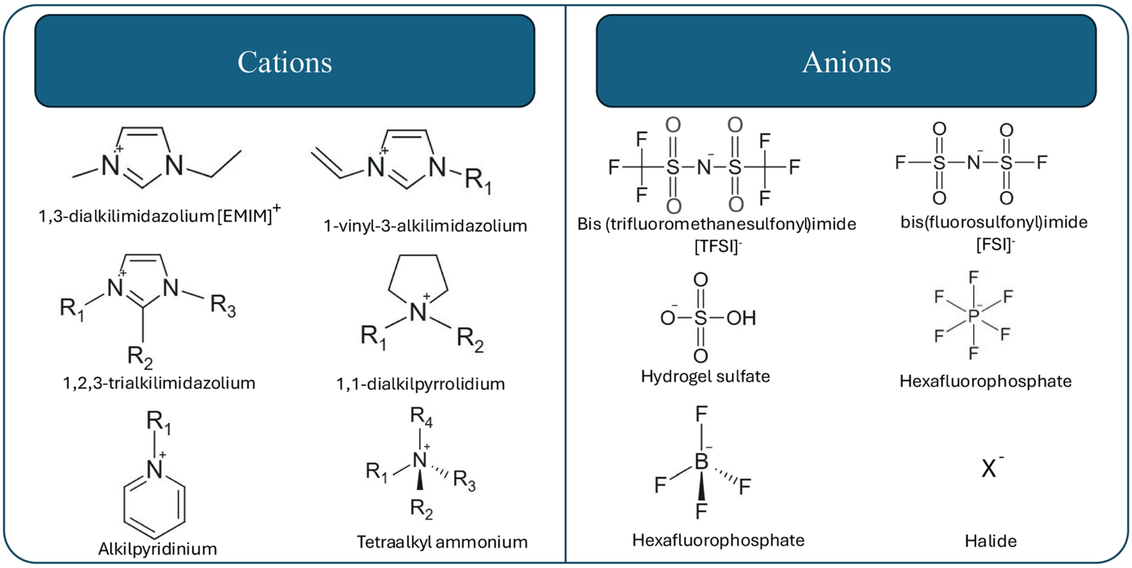

Fig. 5 Chemical structure of cations and anions in common ILs for application in electrolytes.

interactions, and coordination bonding. GPEs with a hydrogen bonded physical network enhance ionic conductivity, mechanical strength, and safety while enabling unique functions such as flexibility and self-healing. Systems like agarose/poly(vinyl alcohol),[122] 2-ureido-4[1*H*]-pyrimidinone (UPy)-based PEO networks,[123] and polyacrylamide/κ-carrageenan double networks demonstrate how reversible hydrogen bonds disrupt crystallinity, provide Li^+ transport channels, and create recoverable structures. These designs make GPEs attractive for high-performance, safe, and durable batteries and supercapacitors.

In coordination polymer gels, the metal ions coordinate with the polymer and create a 3D physical network. These gels are mainly studied in drug delivery, gas storage, and optoelectronic systems.[124–127] One of the major challenges of coordination polymer gels for electrolyte applications is the competition between electrolyte ions and coordination ions (*i.e.*, nodes/crosslinkers) for interaction with the ligand/linker of the network. This interaction can release the coordination ions, which may interfere with Li^+ (or target ion) transport, disrupt the gel network, and/or deteriorate the electrode surface.

GPEs with a physical network can also be formed by self-assembly, *e.g.*, from block copolymers in selective solvent(s).[128] In fact, block copolymers are of significant interest for creating ionogels due to their ability to self-assemble,[129–131] potentially leading to easily processable and mechanically robust materials. Lodge and coworkers have investigated the preparation, structure, rheology, and applications of ionogels formed from block copolymers and ILs.[132–134] Imaizumi *et al.* synthesized ABA-triblock copolymers, polystyrene-*block*-poly(methyl methacrylate)-*block*-polystyrene (SMS), and prepared ionogels by blending SMS with [C_2mim][TFSI] ionic liquid. The gels had phase-separated polystyrene domains as physical crosslinks and a PMMA-rich phase for ion conduction, achieving relatively high ionic conductivity ($>10^{-3}$ S cm^{-1} at room temperature). These gels, used as electrolytes in ionic polymer actuators, enabled soft bending under low voltage (<3.0 V), with tunable properties controlled by the polystyrene fraction and IL content.[132] He *et al.* demonstrated the self-assembly of poly(styrene-*block*-ethylene oxide-*block*-styrene) (SOS) triblock copolymers in the ionic liquid [BMIM][PF_6], forming transparent ionogels with as little as 5 wt% SOS. These gels maintain high ionic conductivity, exhibit thermal stability up to 100 °C, and possess notable mechanical strength, showcasing their potential for versatile processing and applications.[134]

As another approach for producing non-covalent networks, the dipole–dipole and ion–polymer interactions can be used in systems composed of ionic polymers, *e.g.*, zwitterionic (ZI) polymers. These interactions provide both mechanical strength and conductivity. As an example in this case, the cation group on the ZI polymer backbone can interact with the anion of the electrolyte, thus, reducing Li^+–anion pairing and enhancing ion mobility with the resulting conductivity near 1 mS cm^{-1}.[135,136]

Chemically crosslinked network. In many studies, chemically cross-linked GPEs are *ex situ* prepared as films, which are then placed between the electrodes.[137–140] *Ex situ* GPEs cannot penetrate deep layers of the electrode and partially fill the internal pores of the cathode, making them less effective for reducing battery resistance. In other words, a fraction of the electrode surface interacts with the GPE, leaving a significant portion of the active material unused.[141,142]

In situ preparation of GPEs, which has attracted attention, is done by injecting a liquid precursor into the electrode before polymerization. In this method, the liquid precursor readily penetrates the internal pores of the electrode, enhancing the contact of active materials. This process can address the challenges of *ex situ* GPEs. Polymerization or crosslinking is typically initiated thermally using molecules like azo compounds[143–146] or peroxides[145,147,148] or can be initiated *via* photo-curing.[140,149] It should be noted that *in situ* preparation methods offer significant advantages, including the creation of efficient pathways for metal cation transport within electrodes and a substantial reduction in interfacial resistance due to excellent interfacial contact.[150,151]

To increase the ionic conductivity in GPEs, different methods have been used, *e.g.*, introducing zwitterionic compounds.[152,153] Copolymerizing the polymer with zwitterionic monomers in ionogels helps optimize Li^+ coordination and improves Li^+ transport kinetics. For example, zwitterionic groups introduced by (co)polymerization of 2-methacryloyloxyethyl-phosphorylcholine (MPC) can interact with $TFSI^-$ and form a $TFSI^-$/MPC shell, which coordinates with Li^+. This competitive interaction of $TFSI^-$ and zwitterions with Li^+ decreases the desolvation energy of Li^+, leading to an ionic conductivity of 4.4×10^{-4} S cm^{-1}. Additionally, the coulombic interaction between $TFSI^-$ and zwitterions significantly decreases the reduction stability of $TFSI^-$, promoting *in situ* formation of an LiF-enriched solid electrolyte interphase (SEI) layer on the lithium metal surface.[1]

Classification of GPEs based on ion type. In 1913, Lewis and Keyes revealed that Li is the most electromotive metal.[154] The oxidation of a unit weight of Li releases a large number of coulombs, which means a high gravimetric capacity (3860 mAh g^{-1}).[155] Thus, lithium was commercialized in the battery application by Sony Co. in 1991 for the first time.[156] Since then, Li has been the most important metal in electrolytes production. However, due to the rising demand, limited availability, and uneven distribution of lithium, interest in batteries beyond lithium-ion batteries (LIBs), such as sodium-ion (NIBs) and potassium-ion batteries (KIBs) has grown significantly.[155] Given the abundance of sodium in the Earth's crust and oceans, NIBs are being extensively researched as cost-effective alternatives to LIBs.[157] Some companies have already begun commercializing NIBs, particularly for stationary energy storage applications, such as capturing energy from intermittent renewable sources like solar and wind.[158,159]

KIBs are attracting growing interest due to their low cost compared to lithium, and a reduction potential close to that of lithium, −2.94 V *vs.* standard hydrogen electrode (SHE) for K^+/K compared to −3.04 V *vs.* SHE for Li^+/Li. The magnitude of reduction potential for Na^+/Na is lower, being −2.71 V *vs.* SHE. Like sodium, potassium does not form an alloy with aluminum, allowing the use of an Al current collector for both the anode and cathode. Additionally, K-ion organic electrolytes offer high ionic conductivity and efficient K-ion diffusion, supported by potassium's weaker Lewis acidity and low desolvation energy due to a smaller Stokes radius (3.6 Å) than Li^+ (4.8 Å) and Na^+ (4.6 Å) in propylene carbonate.[160,161]

Multivalent ions such as Mg^{2+}, Ca^{2+}, and Al^{3+} have emerged as promising charge carriers in battery systems, thanks to their small ionic radii, abundance in the Earth's crust, and stable valence states. These ions are seen as key components in the next generation of batteries.[162] Magnesium-ion batteries (MIBs) have recently gained attention for their potential performance, which is expected to be comparable to that of LIBs.[163,164] Magnesium is an ideal material for practical battery anodes due to its low electrochemical equivalence (∼12.15 g eq^{-1}), relatively negative electrode potential (−2.3 V *vs.* SHE), low cost, and enhanced safety compared to lithium. The low mobility of Mg^{2+} cations can be mitigated by GPEs since they facilitate ion transport through polymer segmental motion and solvating domains.[165]

Calcium-ion batteries (CIBs) are a cost-effective, high-power, and environmentally friendly alternative to LIBs. Calcium has a 2+ oxidation state, providing a higher energy density and a higher cell potential. Also, its standard reduction potential is 170 mV above that of lithium. However, CIBs face challenges with low transference numbers for Ca^{2+}, meaning conductivity is mainly due to anion transport. This challenge can be improved by modifying electrolytes, for example by adding Lewis acids to enhance cation mobility, which can be achieved by composition modification.[166–169] Therefore, GPEs can play a critical role in further development of CIBs.

Aluminum, as the third most common element on Earth, is an increasingly promising material for next-generation energy storage devices due to its unique properties. It features a high gravimetric capacity of 2980 mAh g^{-1}, approximately 77% that of lithium, and a high volumetric capacity of 8046 mAh cm^{-3}, nearly four times that of lithium. Aluminum can be repeatedly stripped and deposited without dendrite formation, making it ideal as a metal anode. Moreover, aluminum's intrinsic safety characteristics and favorable electrolyte properties further enhance its appeal. Therefore, aluminum-ion batteries (AIBs) are emerging as a safe, low-cost, and environmentally friendly energy storage option for post-lithium battery technologies.[170–172] Recent advances in room-temperature IL electrolytes have enabled efficient aluminum cycling, supporting the rapid progress of durable AIB technologies.[173] However, AIBs still suffer from issues like moisture sensitivity and corrosivity. Alternatives like deep eutectic solvents and GPEs may address these limitations, aiming to enhance stability for practical applications.[174]

Comparing the conductivity of several GPEs with SPEs in Fig. 6A reveals higher average conductivity in the GPEs (∼3.9×10^{-3} S cm^{-1}) compared to SPEs (∼1.1×10^{-3} S cm^{-1}) for a wide range of transference numbers of 0.2 to 1. The conductivity against tensile strength behavior in Fig. 6B demonstrates a decreasing trend in the conductivity of polymer electrolytes with increasing tensile strength. The straight line suggests the limiting trade-off for GPEs. The data points for a common liquid electrolyte (mixture of EC and DMC) are also provided in this figure, where its tensile strength is considered zero.

Rheological considerations

In LIBs, the continuous cycling of lithiation/delithiation of the electrodes induces cyclic volume change in the electrode, which poses a cyclic strain on the electrolyte sandwiched between the electrodes. Fig. 7 schematically shows the volume change in the electrodes during the charging/discharging process. In the charging process, the Li^+ ions accumulate in the anode and cause volume expansion, while in the discharging process the cathode is expanded. In addition, the lithiation/delithiation

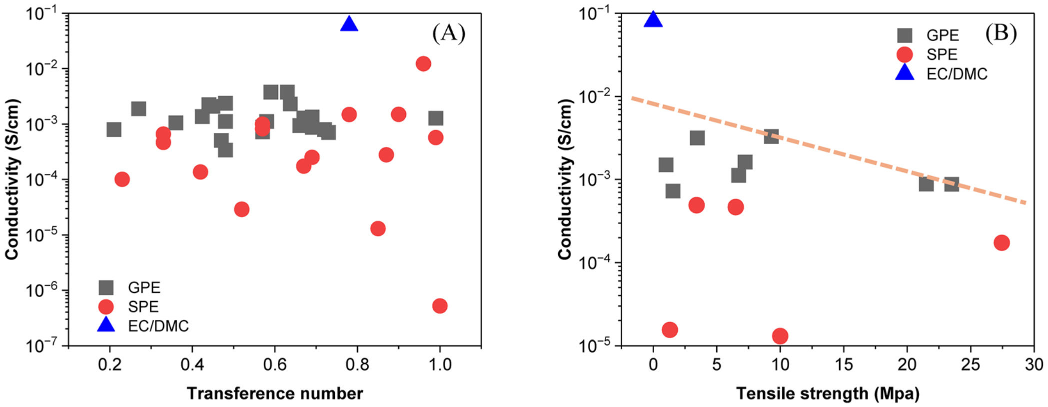

Fig. 6 (A) Conductivity against cation transference number, and (B) conductivity against mechanical strength from recent studies comparing SPEs (red circles) and GPEs (black squares) and an EC/DMC mixture (blue triangle). The straight line suggests a trade-off limit between strength and conductivity in GPEs.[76,78,175–197]

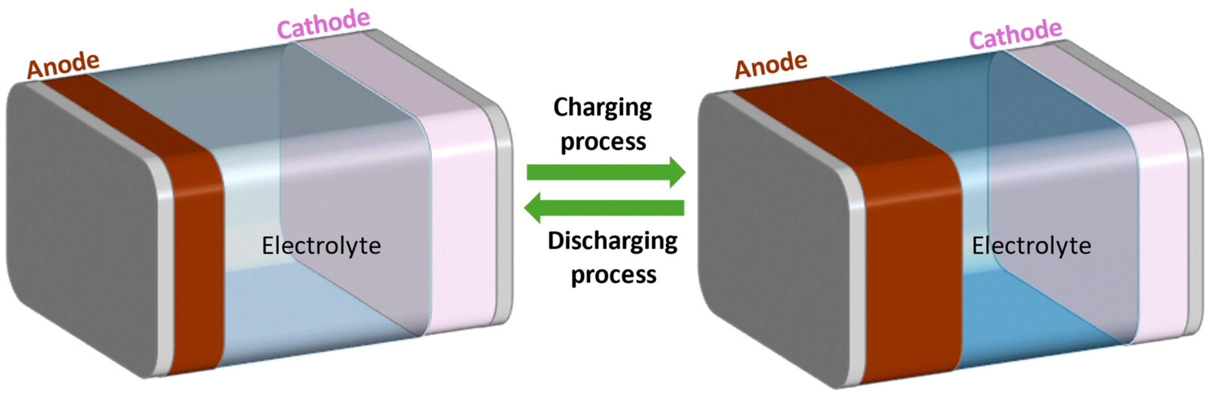

Fig. 7 Schematic illustration of cyclic expansion and contraction of the electrolyte in response to volume changes of the electrodes during charge–discharge cycling. During charging, ion insertion into the anode causes its expansion, while the cathode expands during discharging. Consequently, a net change in electrolyte volume is present during cycling.

process involves heating and cooling of the battery, and thus, electrolyte. This phenomenon is also present in other monovalent- and multivalent-ion batteries. The ability of the electrolyte to keep up with the volume and temperature changes determines the lifetime of the battery.

Table 2 provides some typical values for volume change of electrodes. The volume change in the anode can vary between 0.2% and 270%. For electrodes with a thickness of around 50–100 μm,[198,199] the dilatation of the anode is approximately in the range of 0.1 to 270 μm. Considering the electrolyte with ~100 μm thickness,[200] electrodes can typically impose strain between 0.1% and 270%. The liquid electrolytes accommodate the applied strain by viscous flow, whereas solid-like electrolytes are required to exhibit elastic behavior to respond to electrode volume fluctuations during charge/discharge cycles. Therefore, one of the most important properties of solid electrolytes is a yield strain significantly higher than the imposed electrodes' strain to guarantee elastic behavior and minimum debonding from the plastic deformation of electrolytes. For GPEs and SPEs, the yield strain is around 9–25%[201] and 1.9–14%,[199] respectively. The inorganic electrolytes are brittle with a much lower failure strain, on the order of 0.75%.[202] Therefore, inorganic and SPEs are more prone to mechanical failure and have a shorter cycle life than GPEs.

To minimize cyclic deformation of the electrolyte due to electrode volume change, anodes with zero-strain intercalation are a potential solution. For example, only a slight (0.2%) change in volume caused by lithiation/delithiation is observed in lithium titanium oxide ($Li_4Ti_5O_{12}$, LTO).[206] However, the reported volume changes appear to be in ideal studies rather than practical cases. In addition, LTO suffers from destructive

Table 2 Some common cathodes and anodes with corresponding volume expansion percentages during charging/discharging cycles

	Material	Volume change (%)
Cathode	$LiCoO_2$	2[203]
	$LiMn_2O_4$	3.1[204]
	$LiNiO_2$	9[205]
Anode	Graphite	10[206]
	Si	270[206]
	LTO ($Li_4Ti_5O_{12}$)	0.2[206]

gas generation due to surface reactions.[207] In practical situations, the cyclic strain on an electrolyte is currently inevitable during a battery's lifetime.

For strains below the yield strain of GPEs and SPEs, the cyclic compression/expansion posed on the electrolyte during the charging/discharging process can be simulated by small amplitude oscillatory shear or elongation (SAOS and SAOE, respectively) experiments. SAOE induces change in flow direction and involves aspects of both uniaxial and biaxial elongation. Considering experimental challenges associated with performing elongation flows, SOAE tests are neither straightforward nor common. Since SAOS and SAOE material functions in the linear viscoelastic regime are proportional, SAOS results can be used to simulate the performance of GPEs and SPEs.

In a linear viscoelastic regime, the applied strain is below the yield strain of electrolyte. However, the compression/extension process may still create fatigue failure or aging, which can be categorized as low-cycle fatigue (LCF) or high-cycle fatigue (HCF). LCF occurs under high stress levels, close or beyond yield strain, leading to failure after a relatively small number of cycles. Because the stress exceeds the material's elastic limit, plastic deformation happens during each cycle. In contrast, HCF involves stress levels much lower than yield strain and can endure many more cycles. In this case, the material undergoes mainly elastic deformation, staying below the yield point throughout the cycling.[208] The evolution of dynamic moduli under time sweep tests at constant frequency and strain can be used to study the fatigue and aging of GPEs.

If the volume change in the electrodes imposes strains larger than the yield strain of GPEs and SPEs, the deformation in the electrolyte becomes unrecoverable. Consequently, upon electrode contraction, the electrolyte will not recover back to the original thickness by itself. To investigate such scenarios, large amplitude oscillatory shear (LAOS) studies on the electrolyte are beneficial. Fig. 8 shows typical Lissajous curves for SAOS and LAOS behaviors. In the SAOS, shear and applied strain have linear correlation (Fig. 8A), while for LAOS the correlation is nonlinear (Fig. 8B). Considering the possibility of plastic deformation of electrolytes, a strong adhesion can suppress debonding of electrode and electrolyte, which will be discussed in the next section.

During the charging/discharging process, the volume expansion/contraction of electrodes induce stress in the electrolyte with the mechanical resistance determined by the modulus, which can be approximated by storage modulus and relaxation modulus obtained from rheological experiments. Since the total strain is fixed in this process, the lower the modulus is, the lower the generated stress will be. The associated stored energy (G_{stored}) per unit volume is the area under the stress–strain curve. Based on Fig. 8C, the stored energy in either the charging or discharging stage (*i.e.*, half cycle) can be approximated as:

$$G_{\text{stored}} = \frac{1}{2}\int_{-\varepsilon_0}^{+\varepsilon_0} V\sigma \mathrm{d}\varepsilon \tag{1}$$

In which, σ is the stress, ε is the strain, and V is the electrolyte volume with contact area of A with the electrode and thickness h. Therefore, the stored energy in an electrolyte can be estimated from the electrolyte viscoelastic modulus:

$$\sigma = \varepsilon_0[E'(\omega)\sin(\omega t) + E''(\omega)\cos(\omega t)] \tag{2}$$

where ε_0 is the maximum strain amplitude; E' and E'' are the storage and loss components of complex modulus $E^* = E' + iE''$ in elongation mode, respectively; ω is the frequency; and t is the time. The dynamic shear modulus, $G^* = G' + iG''$, from standard rheological measurements can be used to get the measure of E' and E'' by using complex Poisson's ratio, ϑ^*, as follows:

$$E^* = 2G^*(1 + \vartheta^*) \tag{3}$$

In many practical cases, Poisson's ratio is approximately constant, often assumed to be around 0.3–0.5 for soft materials and 0.5 for incompressible materials including gels.

Another phenomenon to be considered at high charge/discharge rates is the generation of significant Joule heating due to the internal resistance (also known as resistive or ohmic heating). This heat raises the temperature of the battery

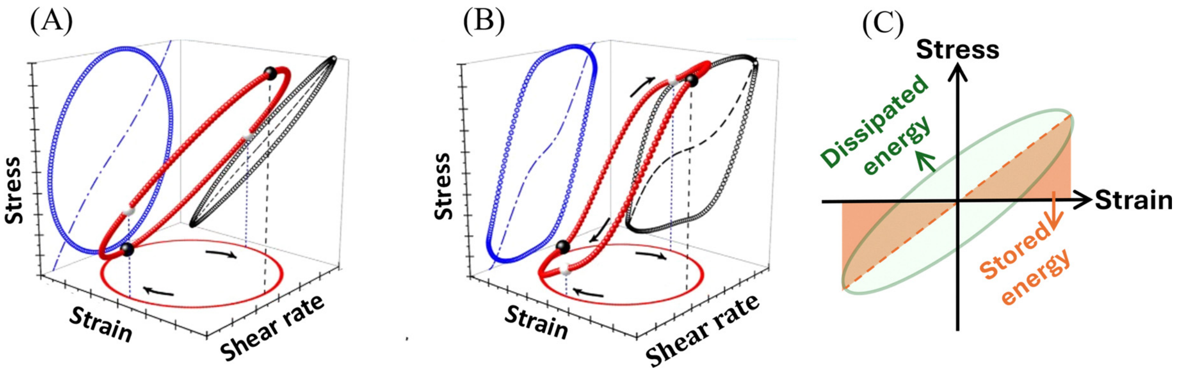

Fig. 8 3D Lissajous curves with projections on the stress–shear rate, stress–strain, and strain–shear rate planes for typical (A) SAOS and (B) LAOS experiments.[209] Reproduced from T. B. Goudoulas, A. Didonaki, S. Pan, E. Fattahi and T. Becker, *Polymers* 2023, **15**(6), 1558, under the terms of the Creative Commons Attribution (CC BY) license. (C) A schematic of the maximum stored energy (hatched area) and dissipated energy (the area inside the curve) in a typical Lissajous curve.

components, causing thermal expansion of internal materials such as the electrodes, electrolyte, and separator. The accumulated expansion of these components leads to a noticeable swelling of the battery cell, which can compromise the mechanical integrity, increase internal pressure, and potentially lead to safety issues such as leakage, debonding of the electrode and electrolyte, or even thermal runaway if not properly managed.[210] The Joule heating has a wide effect on the electrolyte. First, the yield strain in the polymer gels usually decreases by increasing temperature; therefore, their tolerance range for being in the linear region decreases. However, for physical gels, temperature change can cause structure changes and/or sol–gel transition, providing possibilities for rejuvenation if properly exploited. It should be noted that different thermal expansion coefficients for electrodes and electrolyte can induce additional bulk strains on electrolytes upon temperature change.

Another rheological consideration for GPEs and SPEs is stress relaxation behavior. In other words, keeping the volume change for a long time (*i.e.*, battery at rest or disconnected from circuit) induces a force at almost fixed strain on the electrolyte, which may lead to a nonrecoverable deformation in it. Therefore, performing stress relaxation experiments above and below yield strain are beneficial. Alternatively, linear viscoelastic results obtained from SAOS can be converted to stress relaxation behavior using established mathematical methods, such as the Krieger–Kocsis approximation.

Interfacial phenomena at electrode–electrolyte junctions

Interfacial phenomena, such as instability and poor contact, significantly hinder the practical use of lithium metal anodes. Lithium's high reactivity leads to side reactions with organic electrolytes, forming an SEI that acts as a barrier between the electrolyte and the electrode, improving the system's dynamic stability.[211] However, this interphase is typically weak and brittle, which influences the battery's cycling stability.[212,213] For instance, the large volume fluctuations during lithium plating can induce cracking in the SEI, which promotes uneven Li deposition and the growth of dendrites. The repeated breaking and reforming of the SEI layer continuously deteriorates both lithium metal and electrolyte, reducing Coulombic efficiency and eventually leading to battery failure.[214] Therefore, interfacial instability is the primary obstacle to the real-world implementation of lithium metal anodes.[214,215] This is the reason why in conventional LIBs, graphite and $LiMO_2$ compounds (with M being mainly a mixture of Ni, Co, Mn, and/or Al) are used as the anode and cathode, respectively.[206,216]

Interfacial instability in LIBs arises from several factors on both anode and cathode sides. In liquid electrolytes, recent studies of composite graphite electrodes showed that in response to periodic volume changes in the graphite particles, their morphology and the surface films that cover them are very stable in carbonate-based electrolytes, such as EC and cosolvents of DMC, DEC, or EMC and salts of $LiAsF_6$ or $LiPF_6$. However, the surface films may not be able to fully accommodate the volume changes of the graphite particles due to intercalation with lithium. Over time, small-scale reactions occur between the solution and lithiated graphite as solution species pass through the surface films. These reactions create insoluble compounds that thicken the surface films, gradually increasing the electrode's impedance during repeated cycling.[217]

For SPEs, the significant volume change of the anode during charging and discharging can easily lead to detachment at the interface, increased resistance, hindered ion/electron transport, and even dendrite formation.[218] These issues can be hindered by using GPEs instead of SPEs. On the cathode side, SPE decomposition also contributes to high interfacial resistance and cell degradation.[219] Additionally, uneven lithium transport can cause localized reactions, stress accumulation, and microcracks in active materials, while metal ion diffusion from the cathode into the electrolyte can further degrade performance.[220] These issues can be hindered by using GPEs instead of SPEs.

GPEs can be used to address the issue of reactivity of lithium metal anodes with electrolyte without deteriorating conductivity.[221] GPEs also provide polymer elasticity to endure the volume expansion of the electrode and the liquid-level wettability to decrease the interface resistance.[221] The adhesion between electrolyte and the electrode plays an important role in the stability and resistance of the interfacial layer. There is a strong correlation between interfacial resistance (R_{int}) and adhesion strength (σ_{adh}): lower R_{int} values correspond to higher adhesion strengths. Therefore, increasing the wettability enhances the interfacial adhesion and interfacial strength, decreasing R_{int}.[222] There are reports showing that by moving from solid electrolytes towards GPEs, the wetting property improves, R_{int} decreases, and Li ion migration across this interface will become easier.[223]

Adhesion can be theoretically characterized by calculating the work of separation for different atomistically represented molecular models of the surfaces in contact. The work of separation (W_{sep}) has been defined by Finnis[224] as the reversible work required to separate an interface into two ideal free surfaces while suppressing plastic and diffusional degrees of freedom. It can be defined unambiguously and can therefore be calculated by theoretical models. The work of separation per unit area can be related to the interfacial tension between phases 1 and 2 (γ_{12}) and the surface free energies per unit area of the individual surfaces (γ_1 and γ_2) by the Dupré equation:[225]

$$W_{\text{sep}} = \gamma_1 + \gamma_2 - \gamma_{12} \tag{4}$$

For specific electrodes, therefore, we need to increase the surface energy of the electrolyte and/or decrease the interfacial tension to increase the W_{sep}. Enhancing interactions between the electrode surface and the solvent can reduce interfacial tension,[226] for example by increasing adsorption and binding energies,[227–229] matching polarities based on the similar

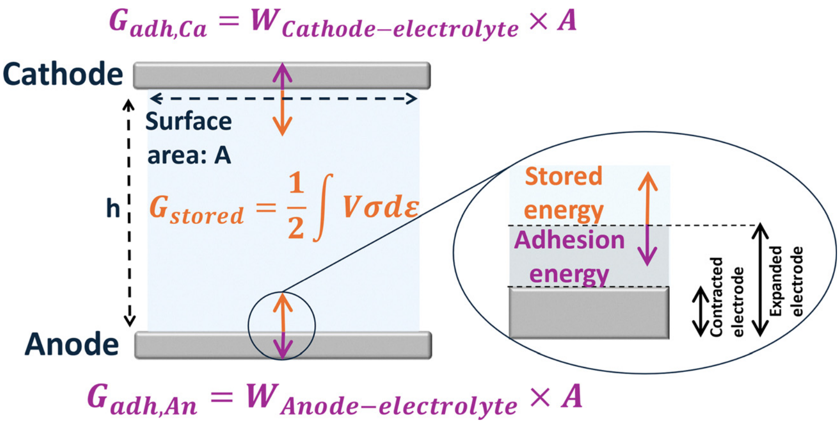

Fig. 9 Competition between elastic energy stored in the electrolyte under cyclic deformation and interfacial adhesion energy governs the mechanical stability of the electrode–electrolyte interface.

polarity compatibility,[230] and promoting hydrogen bonding[231] and electrostatic interaction.[232]

The interfacial tension and the Flory–Huggins interaction parameter (χ) are related:[233]

$$\gamma_{12} = kT(\rho_0\chi)^{\frac{1}{2}}\left[\frac{\beta_A+\beta_B}{2}+\frac{1}{6}\left(\frac{\beta_A-\beta_B}{\beta_A+\beta_B}\right)^2\right] \quad (5)$$

where $\beta_i = \rho_i b_i$, in which ρ_0 is the number average monomer density and b_i is the Kuhn statistical segment length; k is Boltzmann's constant; and T is the temperature.[233] This relationship indicates that systems with higher affinity (lower χ) have lower interfacial tension, meaning higher work of adhesion and more stable interface.

To minimize the electrode–electrolyte separation in electrolytes, especially during the discharging cycle that the electrolyte undergoes extensional deformation, strong molecular interactions are required between the electrode and electrolyte. The presence of functional groups that make GPE more electronegative and can form a solvated structure of lithium ions suppress Li dendrite formation and lead to more stable SEI, diminishing electrode–electrolyte separation.[181]

The adhesion energy between the electrode and electrolyte resists against the separation:

$$G_{adh} = W_{sep} \times A \quad (6)$$

Since the anode and cathode have different chemistries, their adhesion energy is not the same. Thus, as shown in Fig. 9, interfacial stability is governed by the competition between the strain-induced energy per unit volume stored in the electrolyte and the energy cost of breaking adhesive bonds at each interface. Comparing these quantities can provide a predictive criterion for when electrode–electrolyte separation will occur under repetitive volume changes. As the electrolyte is under tension from two sides, half of the stored energy, which can be

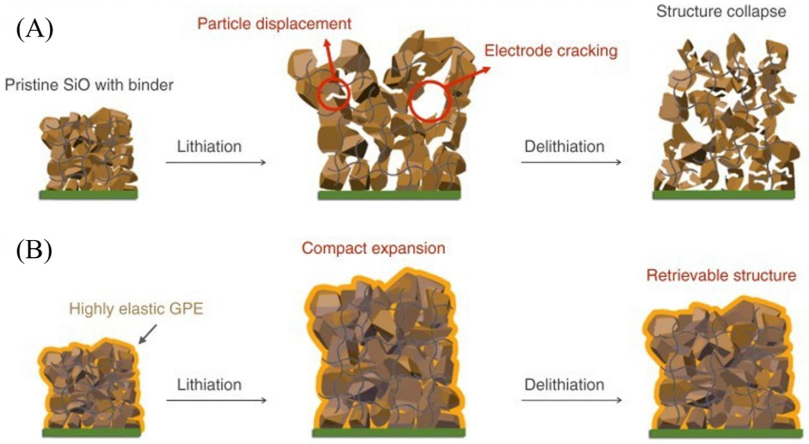

Fig. 10 (A) Conventional SiO electrodes undergo severe structural degradation during lithiation and delithiation due to substantial volume fluctuations of SiO particles, resulting in particle-level and electrode-level cracking. The blue lines indicate the binder network within the electrode. (B) In contrast, the incorporation of a highly elastic GPE provides an integrated electrode structure during cycling. Acting as an intra-electrode cushion (orange), the GPE mitigates thickness expansion and mechanical failure during lithiation and facilitates structural recovery during delithiation.[234] Reproduced from Q. Huang, J. Song, Y. Gao, D. Wang, S. Liu, S. Peng, C. Usher, A. Goliaszewski and D. Wang, *Nat. Commun.* 2019, **10**(1), 5586, under the terms of the Creative Commons Attribution (CC BY) license.

calculated from eqn (1), affects each electrode interface. Therefore, separation takes place when:

$$\frac{1}{2}G_{\text{stored}} > \min\left(G_{\text{adh,An}}, G_{\text{adh,Ca}}\right) \tag{7}$$

where $G_{\text{adh,An}}$ is the adhesion energy between the electrolyte and anode and $G_{\text{adh,Ca}}$ is the adhesion energy between the electrolyte and cathode.

It should be noted that the interfacial adhesion is mainly critical for the two-phase reaction front in the SEI.[232] In addition, the electrode cracking can happen due to several lithiation and delithiation and volumetric changes (Fig. 10A). GPEs can minimize the cracking, consequently improving structural integrity during cycling (Fig. 10B).[234]

Despite the growing interest in multivalent ion batteries, the expansion behavior of electrodes in the presence of divalent ions and their interfacial properties remain underexplored. For MIBs as an example, few materials can reversibly store Mg^{2+} ions, with $Mg_xMo_3S_4$ being the most studied.[235,236] The main challenge is Mg^{2+}'s high charge density, which causes strong electrostatic interactions and hinders ion mobility. To address this issue, there are studies on nanostructured and integration-type electrodes for better performance and cycling stability.[237,238]

Conclusions and future outlooks

In this perspective, gels are defined as biphasic systems comprising a 3D network structure and a liquid phase, which together exhibit solid-like rheological behavior while retaining fluid transport properties. Gels are typically categorized as either physical or chemical. Physical gels are formed through reversible, non-covalent interactions, whereas irreversible, covalent cross-linking is dominant in chemical gels. We further classify GPEs based on their key components and compare the resulting physicochemical and electrochemical properties. A significant challenge in LIBs is electrode expansion during charge/discharge cycles, which imposes cyclic mechanical strain and stress on the electrolyte, potentially leading to structural deterioration. The durability of GPEs under such conditions can be predicted in terms of rheological behavior and in relation to critical interfacial properties, including interfacial tension, wettability, and adhesion strength. Comparing the mechanical energy storage of non-liquid electrolytes with the adhesion energy between the electrolyte and electrode provides insight into the durability of GPEs in charging–discharging cycles.

A key research gap in this field is the lack of systematic studies on how deformation and temperature affect the structure and conductivity of GPEs, implying the need for simultaneous rheological and electrochemical testing. In addition, further studies are required to show how gel networks reorganize during ion transport and cycling. Therefore, we present the following potential future work in this field:

- Perform rheological experiments in SAOS and LAOS regimes while measuring the electrochemical properties to study the effect of cyclic strain and deformation on the electrochemical properties of GPEs.
- Analyze the thermal fatigue within operational temperatures of charging/discharging cycles under cyclic strain and track changes in elastoviscoplastic behavior, gel morphology, mechanical stability, and conductivity.

In addition, there are some remaining research questions that may be addressed in future work:

- What are the differences in ion transport between ordered and disordered structures in GPEs? How can the changes in the internal structure of the interconnected conductive domains affect the ion diffusion?
- How do the non-equilibrium processes (such as repeated cycling deformation and thermal gradients) restructure the physical networks of GPEs or impact network relaxation in chemically crosslinked GPEs?
- Beyond bulk rheology and conductivity, how do different physical and chemical structures change the surface adhesion in GPEs and prevent the Li dendrite formation or affect electrolyte–electrode debonding?
- How does the electrode–electrolyte adhesion affect the interfacial ion transport? What parameters should be considered to produce a GPE with optimal interfacial modulus that enhances facile charge transfer?

Author contributions

F. N. S.: conceptualization, writing – original draft, writing – review & editing. R. F.: conceptualization, writing – review & editing, supervision, funding acquisition.

Conflicts of interest

There are no conflicts to declare.

Data availability

No primary research results, software or code have been included and no new data were generated or analyzed as part of this review.

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