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REVIEW

Polymer electrolyte design strategies for high-performance and safe lithium-ion batteries: Recent developments and future prospects

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Abstract: Although lithium-ion batteries have gained widespread use in high-performance and mobile industries, concerns about their safety due to the low boiling point of their organic liquid electrolyte have posed challenges to their further development. In response, solid polymer electrolytes have emerged as a promising alternative, characterized by low flammability, flexibility, and high safety relative to liquid electrolytes. However, commercialization has been hindered by limitations in Li-ion conductivity and mechanical properties. Recent research efforts have focused on addressing these limitations to improve the performance and safety of polymer-based Li- ion batteries. This review discusses the utilization of polymer materials to enhance battery safety and overcome previous challenges, with a particular emphasis on the design of robust artificial interfaces to increase battery stability. Furthermore, to discuss the prospects for the future of polymer-based battery industries.

Keywords: polymer electrolyte, ion-transport, Li-ion battery, battery safety, solid polymer electrolyte, i-QSE

1 Introduction

Over the past few decades, lithium-ion batteries (LIBs) have become the leading choice for high-performance battery applications due to their compactness, light weight, high voltage (3.7 V), and excellent energy density [1, 2]. They are widely used in portable electronic gadgets, electric vehicles (EVs), and grid energy storage systems. However, the safety issue is a crucial consideration for LIBs. Flammable organic solvents, such as carbonates and ethers, used in traditional non-aqueous liquid electrolytes, pose a significant risk of fire, explosion, and electrolyte leakage, hindering future research on LIB. The dangers are even more significant for post-Li-ion batteries with higher energy density, such as Li-metal, lithium-sulfur (Li-S), and lithium-oxygen (Li-O2) batteries [3–5]. (see Figure 1)

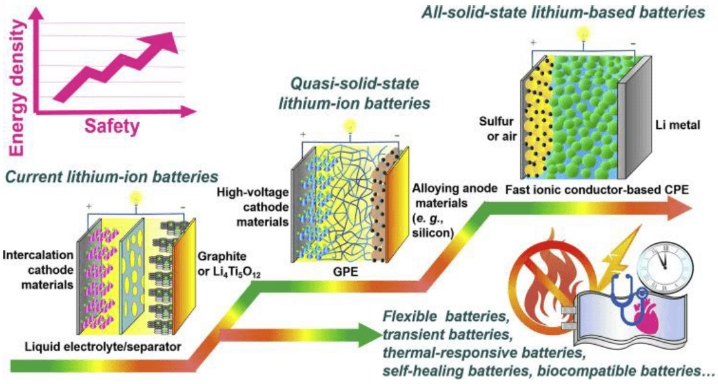


Figure 1 Electrolyte evolution in Li-based batteries [1]

One promising solution to address the safety concerns of Li-based batteries is to replace the traditional liquid electrolytes with polymer electrolytes. Polymer electrolytes can be classified into three types: solvent-free, gel, and composite polymer electrolytes [1,6]. The idea of polymer electrolytes originated from Fenton et al.'s discovery in 1973 that alkali metal salts could be dissolved in polyethylene oxide (PEO) to create conductive complexes [7]. In 1978, Armand et al. developed a PEO-Li salt solid polymer electrolyte (SPE) for Li-based batteries with an ionic conductivity of approximately 10^{-4} S/cm at 40– 60°C [8]. Feuillade et al. introduced organic plasticizers into the polymer-salt binary system in 1975, leading to the development of quasi-solid-state gel polymer electrolytes (GPEs) for Li-based batteries [9]. In 1988, Skaarup et al. made a significant breakthrough by incorporating fast ionic conductor particles, Li_3N, with a high room-temperature ionic conductivity of 10^{-3} S/cm, as fillers into the PEO-$LiCF_3SO_3$ matrix [10].

Polymer electrolytes have advanced rapidly since these pioneering works, as they offer several advantages over traditional liquid electrolytes. These advantages include low flammability, ease of processing, higher tolerance to vibration, shock, and mechanical deformation, better electrode/electrolyte interfacial contact, and compatibility [11]. An ideal polymer electrolyte for Li-based batteries should have high ionic conductivity close to that of liquid electrolytes (10^{-4}-10^{-3} S/cm at room temperature), high Li-ion transference number, excellent thermal and electrochemical stability, and sufficient mechanical strength [1, 12]. High ionic conductivity and high Li-ion transference number can reduce concentration polarization and enhance rate performance. High electrochemical stability (at least 4 V versus Li/Li^+, preferably $\geq$ 4.5 V versus Li/Li^+) enables the use of cathode materials with higher redox potentials, while high thermal stability ensures the safe operation of batteries at elevated temperatures. Good mechanical strength is essential to withstand the volume change of electrode materials during the Li-ion intercalation- deintercalation process and facilitate battery assembly using thin films (15–50 mm) [1, 13–15].

Designing polymer electrolytes that meet these requirements is still a significant challenge. This review paper will summarize recent developments in polymer electrolytes and the corresponding approaches to overcome these challenges, as well as the use of polymers to ensure the safety of batteries.

2 Polymer electrolyte: Issues with liquid electrolytes and possible approaches to overcome difficulties of PEs using polymers

Polymer electrolytes (PEs) became an attractive choice for ion-conducting material in energy storage/conversion devices compared to organic liquid electrolytes because of their high ionic conductivity, enhanced stability features, and safety. PEs are made up of a macromolecule matrix dissolved in a low viscosity, high dielectric constant organic solvent. The development of a covalent link between the polymer backbones and the ionizing groups is the principle of ionic conduction in polymer electrolytes. The polymer's electron donor panel first saves the cation component in the dopant salt, then aids ion separation, resulting in an ionic hopping process that creates ionic conductivity [16].

Table 1 Summary of polymer materials used to enable advanced battery chemistries

Application	Challenges	Solutions	Representative polymer materials
Polymer electrolyte	Low ionic conductivity Low transference number	Low Tg to increase segmental motion [17–19]. Weak cation coordination increases cation diffusivity [20–22]. Anion binding to increase transference number [23].	Modified poly (ethylene oxide) Other polyethers Polycarbonate or polyester
Safety	Thermal runaway	Melting separators for cell shutdown [24] PTC resistors for cell shutdown [25] Flame retardants [26, 27]	Poly(ethylene) Poly(propylene) Fluoropolymers

However, using a Li metal anode with liquid electrolytes frequently results in safety concerns related to the production of uneven metal deposits during recharging, which can lead to the formation of Li dendrites [28, 29]. Due to the thread-like structure between the electrodes, such dendrites can create internal short- circuiting, which can result in fast discharge and uncontrolled thermal runaway [30]. Due to reduced flammability and higher resistance to dendrite formation, the use of lithium ion-conducting polymer electrolytes could eliminate some of the safety issues associated with liquid electrolytes. Moreover, conductivities are typically much lower than those of liquid electrolytes at room temperature, posing a challenge. Polymer electrolytes provide several advantages, including strong ionic conductivity, high energy density, solvent-free state, structural stability, low volatility, wide electrochemical stability window, simplicity of fabrication, and lightweight. Polymer electrolytes are salt solutions dissociated in a polymer host containing an electron donor group. Polymer electrolytes are used as both a separator and an electrolyte in solid-state structures [4, 17, 31]. In terms of form flexibility, size, and weight, polymer electrolytes overcome many of the disadvantages of liquid electrolytes.

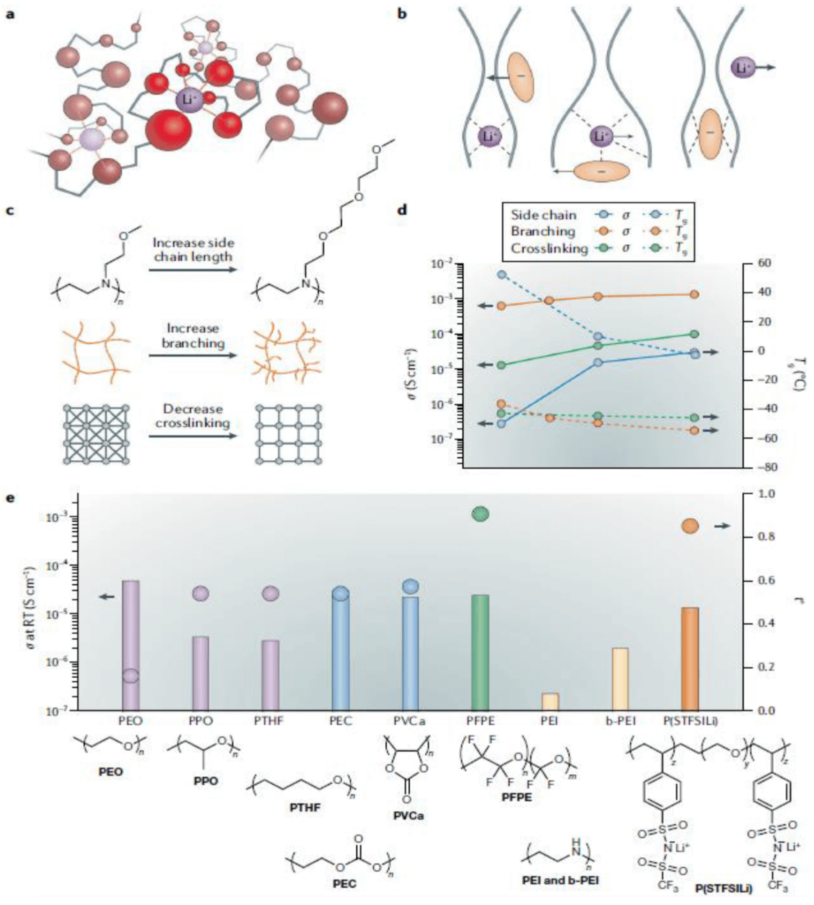
3 Designing polymers for Li-ion transport in PEs

Solid polymer electrolytes (SPEs) have been the focus of intense research, with the promise to allow high- energy-density batteries based on advanced chemistries. Uncontrolled volume expansion, dendrite formation, undesirable side reactions, uncontrolled mass transfer, and thermal runaway are among the issues that solid-state batteries solve. To design effective SPEs, we need to pay attention in the control of the crystallinity, glass transition temperature (T_g), dielectric constant, and solvation site connectivity [4, 16, 17, 31].

3.1 Overview of ion conduction in PEs

PEO was the first polymer electrolyte to dissolve Li ions in 1973 and it is still one of the most researched and commonly used polymer electrolytes today [32, 33]. The dissociation of salt by the polymer backbone and transport of the resultant ionic species by ion hopping or chain segmental motion is the process for ion transport in polymer electrolytes (Figure 2a). In general, an electrolyte's ionic conductivity is a function of the mobility (μ) with which ions may travel through the aforementioned pathways and the concentration of free charge carriers (c):

$$\sigma = \mu C \tag{1}$$



Note: (a) Li^+ hopping from one solvation site to another in poly (ethylene oxide) (PEO). The red dashed lines indicate Li^+ (purple) solvation by O atoms (red) of the PEO polymer, with the transparent purple circles representing potential solvation sites. (b) Schematic showing salt solvation by a polymer backbone. The arrows indicate the motion of the cation or anion, with the thickness of the arrow indicating the relative mobility of each species. (c) Polymer modifications that lead to higher ionic conductivity (σ) include increasing the length of the polymer side chain, increasing branching in a crosslinked polymer, and decreasing the degree of crosslinking. (d) Experimental data show the effects of polymer design on the glass transition temperature (T_g; dashed lines) and σ (solid lines). (e) Comparison of room-temperature (RT) σ and transference number (t^+) of polymer electrolytes with different polymer backbones categorized by functional groups [16]. The polymers are categorized according to the functional groups in the backbone: ethers (purple; poly(ethylene glycol) (PEG), poly(propylene oxide) (PPO) and poly(tetrahydrofuran) (PTHF)), carbonates (blue; poly(ethylene carbonate) (PEC) and poly(vinylene carbonate) (PVC)), fluorinated backbones (green; perfluoropolyether (PFPE)), nitrogen heteroatoms (yellow; poly(ethylenimine) (PEI) and branched- poly(ethylenimine) (b-PEI)) and single-ion conductors (red; lithium poly(4-styrenesulfonyl(trifluoromethylsulfonyl)imide) (P(STFSILi)–block-PEO) [16].

Figure 2 Illustration of ion transport in polymer electrolytes

Given that ion transport through polymer electrolytes is intimately coupled to the motion of the polymer backbone (Figure 2a, 2b), it is not surprising that the empirical Williams–Landel–Ferry relationship developed to describe the viscosity of polymer melts can also be used to describe ion conductivity [34]. These equations for polymer viscosity may be combined to generate the more specific Vogel-Tamman-Fulcher relationship, which is commonly used to explain ion motion in polymer melts [35,36].

$$\sigma = A\exp\left(-\frac{E_a}{R\left(T-T_0\right)}\right) \tag{2}$$

Here, E_a is pseudo-activation energy for ion motion, A is a pre-exponential factor, T is the temperature, T_0 is a reference temperature and R is the gas constant. When discussing ion transport, the reference temperature is commonly considered to be 50K below the T_g, even though it has an arbitrary value in the original Williams–Landel–Ferry equation [37]. According to experiments, T_g -50K is the temperature at which the configurational entropy or free volume of the polymer becomes practically zero [38]. When evaluating the efficacy of polymer electrolytes in Li-ion batteries, the transference number, in addition to total ionic conductivity, is an essential figure of merit. The ratio of cation mobility (μ^+) to total ion movement inside the electrolyte (μ^+ + μ^-, where μ^- is the anion mobility) in response to an electric field is the transference number (t^+) in a binary electrolyte.

$$t^+ = \frac{\mu^+}{\mu^+ + \mu^-} \tag{3}$$

Because transport of the redox-inactive anion causes concentration polarization throughout the cell, which raises the charge overpotential and limits the battery's rate capability, the transference number should ideally be unity [16].

3.2 Effective Li-ion transport in SPEs

Lowering the T_g of the polymer backbone gives a tractable handle to enhance performance when seeking to boost the conductivity of polymer electrolytes (Figure 2c, 2d). Furthermore, except in rare cases, ion conduction in polymers is thought to occur in amorphous areas [39,40]. As a result, increasing the amorphous phase of the polymer while minimizing crystallinity is a primary design aim for polymer electrolytes. A sequence of polymers with similar backbone chemistry but increasing side-chain lengths and therefore varied values of T_g were synthesized as a prominent illustration of the impact of T_g on ionic conductivity (Figure 2c). The equivalent ionic conductivity rose tenfold when the length of the side chain was extended from one to three repetition units [41,42].

Other ways to decrease crystallinity and reduce the T_g of polymer electrolytes include enhancing polymer branching, decreasing the degree of crosslinking (Figure 2c), and synthesizing hyperbranched polymers or bottlebrush polymers [43,44].

The ionic conductivity of an SPE is increased when the T_g of the electrolyte matrix is reduced, but this has no effect on the core Li^+ transport mechanism across the PEO membrane. The degree of salt dissociation and the Li^+ coordination environment in the polymer backbone determine ion conduction within an SPE. The development of 'superionic' polymer conductors, in which ion transport is decoupled from the polymer chain's segmental relaxation, has received a lot of attention. The degree of salt dissociation increases as the dielectric constant of the polymer increases, but Li^+ coordination is often greater in polymers containing Lewis basic functional groups (that is, polymers with larger donor numbers). Despite the lack of rigorous studies on the effect of polymer dielectric constant and donor number on ionic conductivity, it is well understood that ion solvation can be either too strong, in which case the ion is adhered to the polymer backbone and thus immobile, or too weak, in which case the salt does not dissociate [45,46]. The transference number of PEO with lithium bis-(trifluoromethane)-sulfonamide (LiTFSI) is 0.2, indicating that the PEO medium's coordination environment is excessively strong (Figure 2e). Several loosely coordinating polymer electrolytes with greater transference numbers have been studied in an attempt to counteract this strong coordination (Figure 2e). Poly (ethylene carbonate), poly(tetrahydrofuran), polyesters, perfluoropolyether, and polyphosphazene are among the polymer backbones used (Figure 2e). A series of polyborane SPEs containing heteroatoms with Lewis's acidity in the backbone was suggested in a recent computational analysis. The use of heteroatoms allows polyboranes to preferentially solvate TFSI rather than Li^+, resulting in considerably improved Li^+ motility (Figure 2b). Even though the possibility of manufacturing such compounds has yet to be determined, such a technique appears to be promising for overcoming excessive Li^+ coordination in SPEs [47,48].

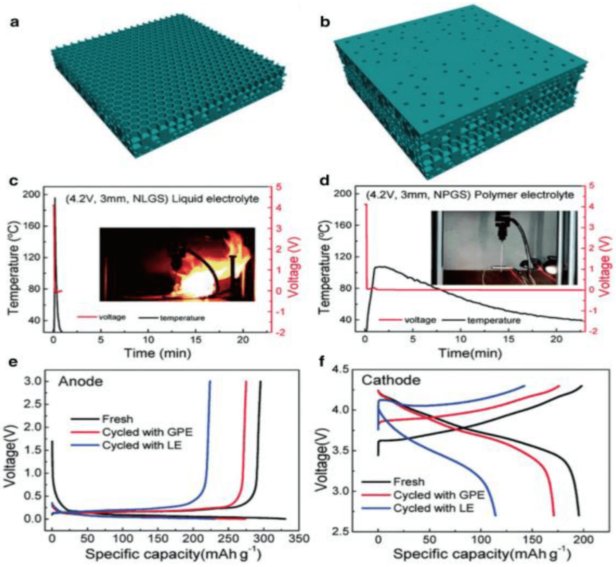
The term "connectivity" has been coined to describe the number of attractive Li^+ solvation sites to which the cation may migrate from its present solvation site. When the connection of solvation sites in the SPE is taken into consideration, differences in ionic conductivity and T_g can be explained. It is advantageous to have uniform solvation sites that are adjacent to each other for effective ion transport in a solid electrolyte. This is why polymer electrolytes like PEO, which are very chemically simple, are so successful. To improve on the low transference number in the traditional PEO electrolyte (t^+ 0.2), many single-ion-conducting SPEs have been created. The anion is covalently linked to the polymer backbone in most of these electrolytes. A block copolymer with a lithium poly(4-styrenesulfonyl(trifluoromethylsulfonyl)imide) (P(STFSILi)) block and a PEO ion-conducting block is the most well-known example of a single-ion conductor (Figure 2e) [49]. The tethered anion is stationary in this scenario, resulting in a transference number close to one. Crosslinking a methacrylate Li salt into a PEO network and incorporating Li-containing nanoparticles into the polymer matrix are two further ways of generating SPEs with high transference numbers.

In the absence of a liquid plasticizer, such attempts resulted in polymer electrolytes with large transference numbers but poor ionic conductivities. The low conductivity is due to a strong ion pairing between Li^+ and the polyanion backbone, as well as the anion's contribution to the total conductivity being reduced [16, 45–48].

To create an effective SPE, an amorphous polymer system with high segmental motion domains is required, as well as a balance between Li^+ solvation and solvation site connectivity. A high transference number is also required in a practical application. Finally, these features should be carefully addressed in terms of mechanical and electrochemical stability, which are two of the most important advantages of switching to solid-state devices.

4 Designing of Polymer/Liquid hybrid system to improve PEs (Both CPEs and GPEs)

Solid polymer electrolytes (SPEs), which offer great flexibility and security guarantee, have grown in popularity in recent years and are being evaluated as viable alternatives to traditional organic liquid electrolytes. SPEs' lower room-temperature ionic conductivity, on the other hand, is a disadvantage that limits their growth in the field of solid-state lithium batteries [50, 51]. To address this, hybrid systems based on polymer/liquid electrolytes have been investigated in recent years and have shown to be capable of balancing the features of SPEs and standard organic liquid electrolytes. Because of their ability to provide superior interfacial contact between the cathode and anode, as well as having high ionic conductivity and flexibility, these polymer/liquid hybrid electrolytes containing certain amounts of liquid ingredients in a polymer matrix have shown increasing promise in the application of electrochemical energy harvest devices. Furthermore, in solid-state lithium batteries, polymer/liquid hybrid electrolytes may build a robust protective layer, ensuring greater safety performance than standard organic liquid electrolytes.



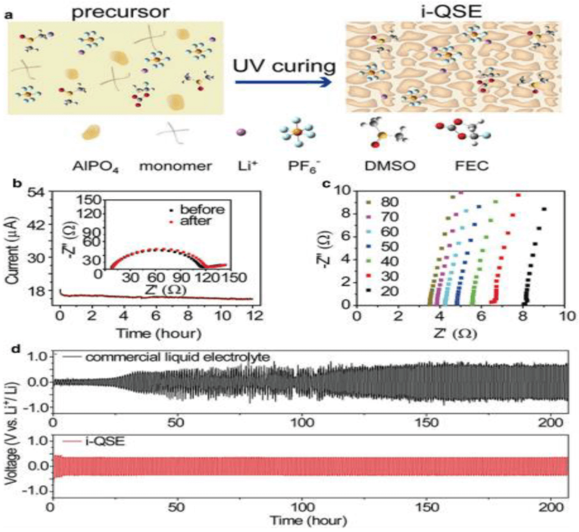
Note: (a) 3D architecture of the PVDF-HFP polymer membrane; (b) porous gel polymer electrolyte with a combination of two membranes as one integrated separator (c); (d) An examination of the safety performance of NLGS and NPGS batteries by nail penetration testing; (e) Specific capacity of the graphite anode and f). NCA cathode before and after 280 cycles at 0.5C charge/1C discharge and 45°C. [52, 53]

Figure 3 Designing of polymer/liquid hybrid system

Furthermore, in polymer/liquid hybrid electrolytes, the volatility and leakage characteristics of organic liquid electrolytes may be effectively prevented, while thermal stability and safety performance can be improved. Zhang et al. [52], for example, developed a honeycomb-like polymer matrix based on poly (vinylidene difluoride-hexafluoropropylene) (PVDF-HFP) that had a high electrolyte absorption of 86.2 percent and outstanding thermal stability up to 350 °C (Figure 3a, 3b). In another study, Li et al. [53] used a radical polymerization synthetic route to synthesize a pentaerythritol tetra acrylate (PETEA)-based polymer/liquid hybrid electrolyte and performed nail penetration tests to investigate the safety performance in which they built a pouch battery using the synthesized PETEA-based electrolyte as well as a pouch battery using a commercially available liquid electrolyte for comparison (Figure 3c, 3d).

During the nail piercing test, the liquid electrolyte battery underwent intense combustion, but the polymer/liquid hybrid electrolyte pouch battery produced far less gas. Furthermore, Li et al. [53] reported that the anode and cathode of the polymer/liquid hybrid electrolyte-based battery in their study could hold higher capacity retentions (Figure 3e, 3f) and that the prepared polymer/liquid hybrid electrolyte had improved safety performances, higher ionic conductivities, and better cycling stabilities when compared to the liquid electrolyte, indicating the construction of a stable interface layer using the liquid gradient within the battery.

In another study, drawing lessons from 'Soggy sand electrolytes [54], Zeng et al. [55] reported an infiltrated quasi-solid electrolyte (i-QSE) with nano-sized AlPO4 interspersed throughout the interconnected polymer network (Figure 4a).



Note: (a) Schematic illustration of the preparation of i-QSE; b). DC polarization with a 10mV amplitude in the Li symmetric cell and impedance curves before and after polarization (insert in Figure (b); (c) Impedance spectra of the i-QSE versus temperature; (d). Voltage profiles versus time of Li symmetric cells in a liquid electrolyte (up panel) and i-QSE (down panel) at a current density of 4 mA cm^{-2} for 2 mAh cm^{-2} [50, 55].

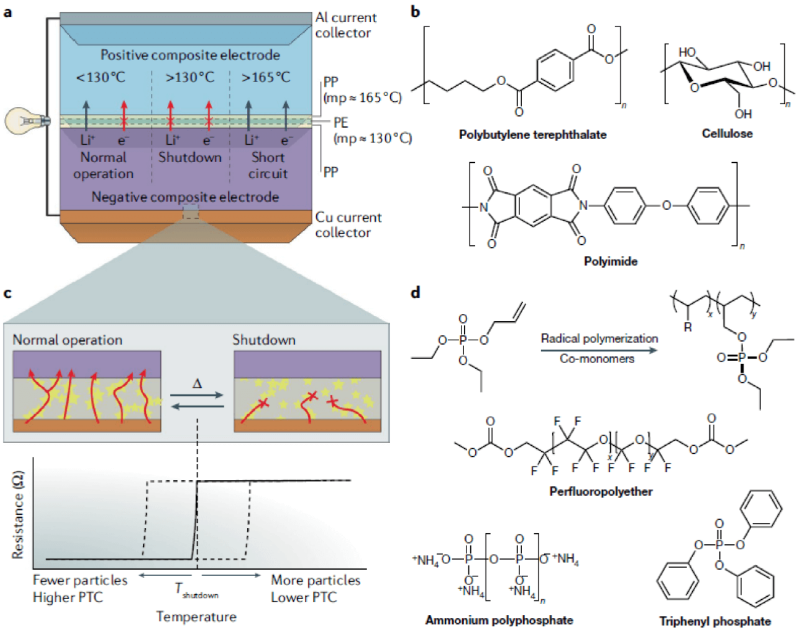
Figure 4 Schematic illustration of the preparation and characterization of i-QSE

This concentrated electrolyte enclosed in an infiltrated structure was discovered to be capable of promoting interface wetting and increasing electrode-polymer electrolyte adhesion. The infiltrating structure was also

shown to help prevent lithium salt depletion and significantly reduce uneven Li electrodeposition, resulting in improved Li$^+$ transport and a decreased activation energy of 0.12 eV. (Figure 4b, 4c). Because of these advantages, a Li symmetric battery built using the i-QSE displayed higher cycle stability at elevated temperatures (55°C) as compared to commercial liquid electrolytes (Figure 4d). These novel interface- improved methods using the in-situ generations of safeguards for Li anodes shed light on promising novel synthesis methods from an interface-engineering level [50, 51].

5 Polymers for safety in batteries

Even with today's technology, unacceptably catastrophic battery failures are prevalent, and the ramifications of battery failures are enormous in terms of user safety and profitability. As a result, it's critical to provide better safety systems for new battery chemistry. Polymers, especially through their thermos- responsive qualities or as non-flammable device components, promise to play a significant role in improving battery safety. Modern Li-ion batteries rely on thermos-responsive polymers for their safety mechanism. Polymer melting and thermal expansion are the two most studied temperature-responsive features in the development of safe batteries [56]. Modifications to the monomer chemistry or polymer design can be used to manipulate these thermal characteristics. The melting temperature of a polymer can be increased by increasing the strength of the ability of the polymer to pack into crystals (for example, by reducing chain flexibility, increasing symmetry, or decreasing branching) [57].



Note: (a) Diagram of a standard cell with a tri-layer polyolefin separator. Shutdown occurs if the polyethylene (PE) layer melts, but a short circuit occurs upon melting of the polypropylene (PP) layers; b) Chemical structures of polymers are used to fabricate separators with high thermal stability; (c) Reversible operation of a thermos responsive polymer composite (comprising PE and graphene-coated Ni microparticles) applied between the current collector and anode. The transition from normal operation to shut down above the shutdown temperature (T shutdown) owing to a large increase in resistance is shown; (d) Chemical structures of flame-retardant polymers used as electrolytes and binders [16, 58].

Figure 5 Polymer materials for improved battery safety

Cell-guard separators have a three-layer structure comprising polypropylene, polyethylene, and polypropylene. Ion transport through the battery is blocked if the battery temperature exceeds the melting point of the polyethylene layer, preventing thermal runaway (Figure 5a). One safety risk with this process is that if the battery warms up to 165°C, the isotactic polypropylene layer would melt as well, resulting in a battery short circuit. Although this feature improves safety, high-thermal-stability polymer separators have also been created to help avoid short-circuiting that happens when polypropylene is melted (Figure 5b) [59, 60]. In addition to the melting separator, Commercial cells often include an external resistor made of materials with a positive thermal expansion coefficient (PTC), which inhibits current flow when heated and prevents thermal runaway [56]. The rate at which thermal runaway occurs, on the other hand, reduces the efficacy of these external devices. The coefficient of thermal expansion in polymers is proportional to the amorphous percentage of the polymer and inversely proportional to T_g [58]. A thermos-responsive polymer composite based on polyethylene and graphene-coated nickel microparticles was designed to be applied directly to the current collector of the cell to improve upon external PTC resistors (Figure 5c). When polyethylene expands, the particles migrate apart from each other and the percolated electron transport channels become disconnected. The shutdown temperature may be adjusted between 50 and 100 degrees Celsius by adjusting the number of nickel particles in the composite and the thermal expansion coefficient of the polymer matrix. Furthermore, the battery function restarts once the polymer matrix cools. Incorporating a reversible safety switch in Li-ion batteries is a desirable feature. Polymer solutions' thermal responsiveness can also be employed to improve the safety of Li-ion battery operation. Many solutions exhibit lower critical solution temperature (LCST) behavior when they begin to phase separate. Because of the negative entropy of mixing in polymer solutions, the interaction becomes more complex as the temperature rises. Thermal runaway in Li-ion batteries has been mitigated using the LCST behavior of electrolytes including poly (benzyl methacrylate) in an ionic liquid and methylcellulose in an aqueous solution [61]. When heated, the polymer in these solutions separates from the solvent and forms an insulating phase, or the electrolyte gels, reducing ionic conductivity in both cases. Despite these attempts, taking advantage of thermos-responsive polymer phase separation does not result in the complete shutdown of the battery. When a solution starts to phase separate, it often has a lower critical solution temperature (LCST). The interaction becomes more complicated as the temperature rises because of the negative entropy of mixing in polymer solutions. The LCST behavior of electrolytes such as poly (benzyl methacrylate) in an ionic liquid and methylcellulose in an aqueous solution has been used to limit thermal runaway in Li-ion batteries. When these solutions are heated, the polymer separates from the solvent and forms an insulating phase, or electrolyte gels, lowering ionic conductivity in both cases. Despite these efforts, using thermos-responsive polymer phase separation to shut down the battery does not completely shut it down [62–64].

6 Conclusions and future perspectives

Even though lithium-ion batteries have long been marketed, the use of liquid electrolytes has several drawbacks, including poor safety and unpredictable electrochemical performance, which severely restricts their development and application. Polymer electrolyte in lithium-ion batteries has recently attracted a lot of attention due to its low flammability, outstanding flexibility, excellent thermal stability, and high safety. In this study, I want to evaluate current developments in polymer-based composite electrolytes such as the polymer/liquid hybris system and i-QSE, as well as fundamental understandings of ionic conductivity processes for solid polymer electrolytes. Even though much research has been devoted to polymer-based electrolytes, certain basic challenges must be resolved quickly before commercialization. For example, recent research on improving ionic conductivity and transport number is going on but it's not enough because the solid polymer electrolyte's ionic conductivity still differs by several orders of magnitude from its liquid counterpart; many polymer-based solid electrolytes have high ionic conductivity at high temperatures, but it drops dramatically at lower temperatures; the conductivity mechanism needs to be clarified to expedite further research. When attaining high ionic conductivity, the polymer/liquid-solid electrolyte always produces a loss in mechanical characteristics, which poses significant safety risks. SPEs with rapid ion conductors have outperformed all other forms of polymer-based composite solid electrolytes and are the direction of commercial solid electrolyte development. The following factors of focusing on solid electrolytes in future advancements were advocated. To begin, employing a materials genome database to evaluate, guide, and develop composite materials can help to increase efficiency and reduce costs. Material calculations aid in gaining a comprehensive knowledge of the material. Material computations can be used to simulate and analyze the related ionic process. Second, most battery materials, such as electrodes, electrolytes, and SEI coatings, are electron beam sensitive and difficult to view with traditional transmission electron microscopy (TEM). Finally, the challenge of developing polymer electrolytes with excellent conductivities at low temperatures requires further study. To compete with their liquid counterparts, commercial polymer electrolytes must have good ionic conductivity at ambient temperature, be safe, and be easy to produce.

Conflicts of interest

The authors declare that there are no conflicts to declare.

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