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PERSPECTIVE

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Tailoring Solid-State Electrolytes via Coordination Chemistry Design for Lithium Batteries Operating in Subzero Environments

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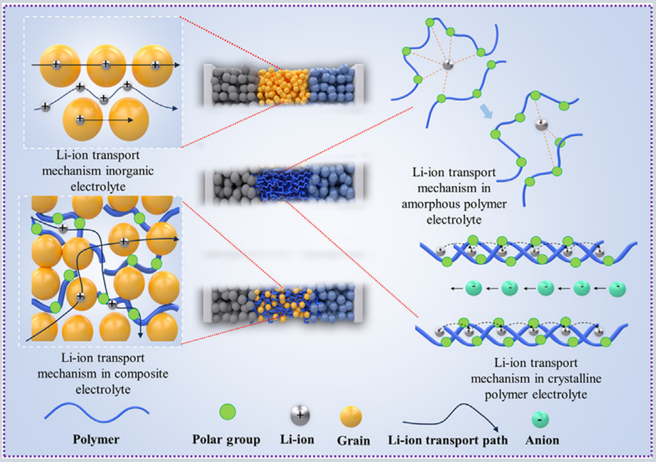
Solid-state electrolytes (SSEs) are highly promising constituents of high-energy-density and high-safety batteries. The remarkable electrochemical performance of solid-state lithium batteries' (SSLBs) at moderate and high temperatures has been demonstrated in extensive research. However, their performance drastically deteriorates in cold environments, and the underlying process of this deterioration is still unknown. In this perspective on the state of research in this area, we analyze ion transport kinetics for SSLBs and highlight their difficulties at low temperatures. Through an analysis of microscopic kinetic processes, such as interfacial charge transfer resistance and the lithium-ion migration mechanism within solid-state electrolyte (SEs), we identified specific requirements and the critical challenges of Li-ion transport, interfaces, and SEs for low-temperature SSLBs. Numerous tactics, comprising interfacial engineering, compositional doping, and flexible composite electrolytes, have been suggested. Improved comprehension and implementation of these solutions may open the door to lithium battery technologies that are more robust, efficient, and temperature-resilient.



Keywords: lithium battery, solid-state electrolyte, low-temperature, ionic conductivity, activation energy

Introduction

In pursuit of high-energy-density lithium batteries (LBs), strenuous efforts have been made to tailor solid-state electrolytes (SSEs) due to their high thermal stability and safety. This is in contrast to the thermal instability, volatility, and inherent flammability of liquid electrolytes, which are major safety concerns.[1–4] However, despite significant advancements in SSEs, a problem has arisen concerning LBs' decayed performance at low temperatures, which has been a major stumbling block in the application of LBs at high latitudes and high altitudes and in specific defense and space applications.[5] For instance, at −40 °C,

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commercial LBs can only provide 10% of their room-temperature capacity. LBs undergo inevitable performance degradation when operated cryogenically (typically below 0 °C, due to capacity fading, cycling instability, and parasite reactions).[6,7] Additionally, the tendency for detrimental Li deposition causes safety and temperature-related issues.[8,9] The use of LBs is restricted in cold conditions due to performance decay and safety concerns.

To address the issue of performance degradation in LBs at low temperatures, it is crucial to thoroughly examine the fundamental steps of lithium-ion (Li^+) transportation during the charge and discharge processes. It has been considered that at low temperatures, the sluggish kinetics of Li^+ is the perilous aspect that restricts the electrochemical performance of batteries. Therefore, prior to devising effective strategies to enhance the performance of solid-state batteries (SSBs) at low temperatures, it is essential to comprehend the kinetic processes involved. A simple classification of these processes can be made as follows: firstly, Li^+ migration within the solid electrolyte (SE); secondly, an electrode/SE interface involving charge transfer; and finally, Li^+ migration within the bulk electrode.[10,11]

The kinetic processes of SSBs must be also discussed in detail in order to understand their complexity. In SSBs, a number of steps are involved in the interphase kinetic process, which includes: (1) the extrication of Li^+ from the sublattice or coordination environment of SE, (2) the transportation of Li^+ ions through the interphase, and (3) the charge transfer at the electrode surface. Finally, the establishment of an interfacial charge transfer process by these steps jointly.[10] Hence, the SSBs fabricated by inorganic SEs have the absence of the energy-consuming desolvation process of Li^+. However, it is unclear whether a similar undetected process occurs at the SE interface and whether Li^+ transport across this interface is subject to the same energy barrier. Also, LBs should have different properties from mosaic-type solid electrolyte interphases with inorganic and organic hybrid components in terms of physicochemical properties. Consequently, it is unclear how an all-inorganic interphase impacts Li^+ transport and follow-up charge transfer.

Herein, this perspective summarizes the challenges of low-temperature SEs and points out how to tailor strategies for the SEs in terms of material design [such as different kinds of inorganic SEs and solid polymer electrolytes (SPEs)] to mitigate ion kinetic problems, which are responsible for better electrochemical performance in low-temperature environments. To achieve the effective, enhanced performance of solid-state lithium batteries (SSLBs) at low temperatures, we also propose that the following essential requirements for their design should be met regarding the materials and chemistry of the battery: (1) To attain enhanced ion transport in SE at extremely low temperature, high ionic conductivity of SE should be maintained. (2) Good interfacial contact should be established by maintaining low interface resistance. (3) Uniform ion transport should be maintained by tailoring the electrolyte materials with higher ionic conductivity at low temperatures. (4) Decent ionic diffusivity of electrode should be upheld. Design strategies for materials to satisfy constraints of low-temperature SSLBs are proposed by tailoring SE via interface engineering.

Overview of ion conduction mechanism in solid state electrolytes

It is critical to understand the ion conduction mechanisms of SSEs in order to boost the performance of SSLBs at low temperatures (Figure 1). For instance, as a result of the crystalline or amorphous structure of SSEs, ions are transported primarily through vacancies, interstitial sites, and grain boundaries.[12] A key factor in ion conduction is the mobility of cations, typically lithium ions (Li^+), which are enabled by diffusion or hops between available sites.[13–15] An inorganic SE (e.g., oxide, halide, sulfide) with an immobile sublattice is carried by Li^+. Li^+ ions are the sole charge carriers due to the immobile anion sublattice, resulting in a Li^+ transference number of 1.[16]

There are two types of Li^+ migration in polycrystalline inorganic SEs (e.g., $Li_{10}GeP_2S_{12}$), namely the migration of Li^+ within the bulk/grain assembly and migration through the grain boundary structure (Figure 1i). From a macroscopic perspective, crystalline inorganic SEs satisfy the Arrhenius equation for ion conductivity as follows:

$$\sigma = \frac{\sigma_0}{T} e^{-\frac{E_a}{K_B}} T \tag{1}$$

where, E_a, σ_0, σ, T, and K_B stand for activation energy, pre-exponential factor, ionic conductivity, absolute temperature, and Boltzmann constant, respectively.

We can deduce from the above equation that the sensitivity of ionic conductivity is attributed to activation energy (E_a) at varied temperatures. Furthermore, according to these insights, E_a is influenced by multiple scale factors,[17,18] including lattice dynamics, structural disorder, and frustration, bottlenecks, lattice volume, migration modes, interactions of charges, lattice defects, and grain boundary effects on a crystal structure level. For instance, Hogrefe and his team[19] describe how limited lattice vibrations at low temperatures affect the Li transportation in crystalline argyrodites. Currently, tactics for reducing activation energy may only apply to certain materials or scales. A specific polyanion has been observed to exhibit rotational dynamics that promote ion hopping.[20] Due to these critical characteristics of crystalline inorganic SEs, it is more advisable to govern a suitable approach than to drop the activation energy.

The Li^+ migration in amorphous inorganic SEs, for instance, Li_2S-P_2S_5 glass, involves only bulk transport, which can be rewritten as the Vogel–Tammann–Fulcher (VTF) equation.[11] An amorphous inorganic SE's ionic conductivity also determines the VTF equation, which is as follows:

$$\sigma = AT^{-\frac{1}{2}} e^{-\frac{E_a}{T - T_0}} \tag{2}$$

where E_a, A, T, and T_0 stand for activation energy, pre-exponential factor, ionic conductivity, absolute temperature, and glass transition temperature, respectively. Although the Arrhenius relationship can be approximated within a temperature range much lower than the glass transition temperature of amorphous SEs (e.g., >2000 °C), the temperature-dependent relationship cannot be approximated outside this range.[21] Furthermore, Chen et al.[22] describe how disorder-induced diffusion in amorphous

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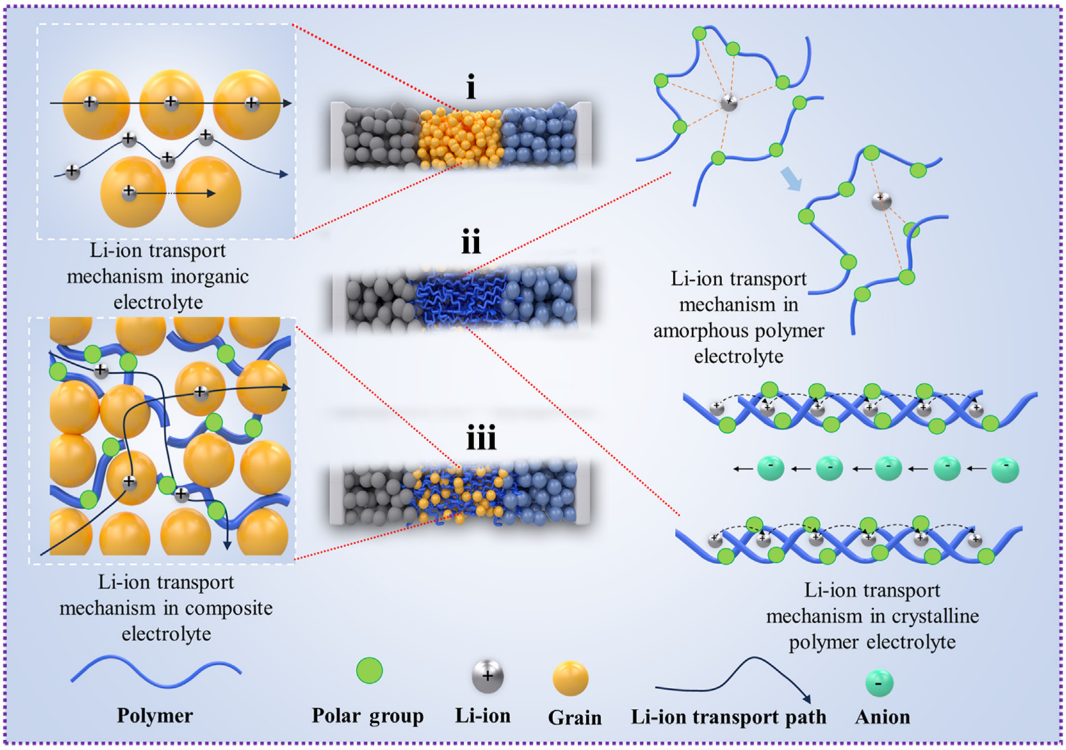

Figure 1 | *Ion-transport mechanism of Li-ion in solid state electrolyte: (i) inorganic solid electrolyte, (ii) SPE, and (iii) composite SE.*

Li_3PS_4 enhances Li-ion conductivity at room temperature and below, driven by dynamic "soft" hopping sites identified through deep learning simulations.

A significant feature of polymer SEs is that Li^+ can partially exist in weak solvation apart from interactions with polar groups in the chain segments of polymer SEs [e.g., polyethylene oxide (PEO)-based SEs], but its transference number is relatively low since anions dissociated from lithium salts tend to move.[23] If a polymer SE is homogeneous and isotropic, Li^+ motion can also be simplified into bulk transport. The ionic conductivity of polymer SEs with low glass transition temperatures also corresponds to the VTF equation because they belong to the amorphous system. A reduction in activation energy can be achieved by reducing the interchain interaction between polymers, as well as weakening the interaction between the polymer's polar groups and anions.[24] When the Li^+ migrates into composite SEs, such as those containing polymer SE and inorganic filler, it becomes more complicated, involving transport through bulk polymer SEs and inorganic fillers (if they are ion conductive) and their interfaces.[25,26]

Design of solid-state electrolyte at low temperature

The prerequisite for low-temperature operation of batteries is the maintainence of high ionic conductivity. The SE has advantages over a wide range of temperatures over organic liquid electrolytes (OLE) which have freezing challenges at subzero temperatures, Although, in recent decades a great deal of effort has been devoted to raising the ionic conductivity of SEs at mild temperatures, very little attention has been given to the question of how to achieve satisfactory levels of ionic conductivity at low temperatures.[27,28]

Inorganic SEs have better ionic conductivity and stability than traditional liquid or polymer electrolytes, which are crucial to the performance of low-temperature LBs.[29] Under low temperatures, conventional liquid electrolytes lose their ionic conductivity and become highly viscous, which limits their ability to transport lithium ions.[30] On the other hand, inorganic SEs, such as ceramics (usually sulfides and garnets), uphold a higher ionic conductivity at subzero temperatures, which allows for better performance in cold atmospheres.[31] Additionally, inorganic SEs provide a broad electrochemical stability window, making it possible to pair with high energy-density materials such as lithium metal anodes, without being concerned about safety issues associated with liquid electrolytes.[32,33] Due to their efficiency at low temperatures, they are crucial to the expansion of LB applications.

To develop the most feasible approach to build better-suited inorganic SEs, here, we address the limitations of inorganic SEs and various strategies associated with required innovative modification approaches (Figure 2). The literature describing how grain boundaries affect the performance of Li-ion batteries is extensive. Pesci and his team[34] elaborate that in $Li_7La_3Zr_2O_{12}$ (LLZO), total impedance below 0 °C is governed by grain boundary resistance; hence, obstruction in ion transport is caused by defect scattering and reduced lattice dynamics. Similarly, Yu and his coworker[35] described how grain boundary segregation of impurities, like Al^{3+} in LLZO, exacerbates this effect by creating potential wells that trap mobile ions, further lowering

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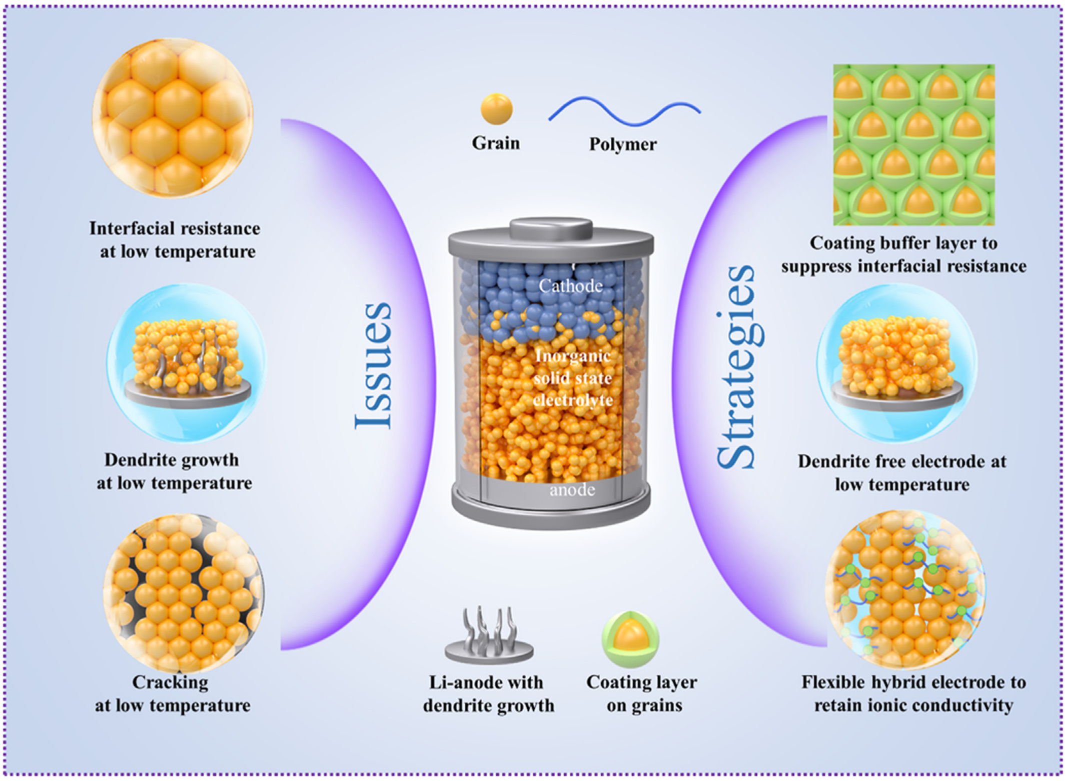

Figure 2 | *Limitations and strategies of inorganic SSE at low-temperature LBs.*

conductivity. Furthermore, inorganic solid-state electrolytes (ISEs) have grain boundaries which are attributed to their polycrystalline nature and hence are typically associated with multiple crystals within them. When materials are sintered or pressed at suboptimal temperatures during the manufacturing process, such as at low temperatures, these boundaries are formed.[36,37] The grain growth can be limited, resulting in smaller grains with more grain boundaries, leading to reduced mobility of ions at low temperatures. Ion transport across these grain boundaries is less efficient than ion transport through the grain interior, which causes these grains to act as barriers for ion conduction. Additionally, the structural mismatch between adjacent grains and crystal orientation increases the resistance at these boundaries, further inhibiting ion diffusion. As grain boundaries play a critical role in SSEs used in low-temperature LBs, they pose a significant challenge due to their diminished ionic conductivity, which adversely affects battery performance at such low temperatures.[38–40] It is, therefore, essential to understand and control grain boundary formation in order to improve the efficiency of SSEs at low temperatures.

There has been a variety of approaches developed to deal with this problem. One of these is to modify its microstructure by reducing grain boundary density through sintering, resulting in larger and more uniform grains with fewer interfaces to cross.[41] Furthermore, doping SEs with foreign cations (such as Al^{3+}, Ta^{5+}, or Nb^{5+}) can reduce ionic and electronic resistance by stabilizing the lattice and enhancing grain boundary diffusion.[42,43] By modifying local chemistry, the ion mobility can be improved at lower temperatures. Similarly, partial substitution can improve grain boundary conductivity by altering anion composition (e.g., substituting sulfur for oxygen). It is also possible to enhance ionic conductivity by coating or modifying grain boundaries with highly conductive secondary phases, such as lithium-rich materials. A composite electrolyte can also be used to create interfaces that can transport ions more efficiently by combining different materials.[44,45] By combining these techniques, grain boundary resistance can be reduced, and inorganic SEs perform better, especially in low-temperature environments where ion mobility is already limited.

The development of lithium dendrites in ISEs is another substantial threat faced by LBs operating at low temperatures. For instance, dendrite growth occurs in SSEs, such as LLZO, regardless of their extraordinary mechanical strength, along defects or grain boundaries, as revealed by Zhang and his team[46] by using molecular simulations. This article describes that lithium dendrites infiltrate SSEs via stress-induced cracking at the dendrite tip, which is governed by electronic conductivity and electrochemical degradation rather than transmission alone. Dendrites are needle-like structures formed by lithium metals during repeated charging and discharging cycles. The growth of dendrites is attributed to the uneven deposition of lithium ions, which caused a high interfacial resistance and reduced mobility at lower temperatures. SEs made of inorganic materials, such as ceramics and sulfides, are generally strong mechanically, but their reduced ionic conductivity and brittleness can exacerbate dendrite formation at low temperatures. Due to the mismatch

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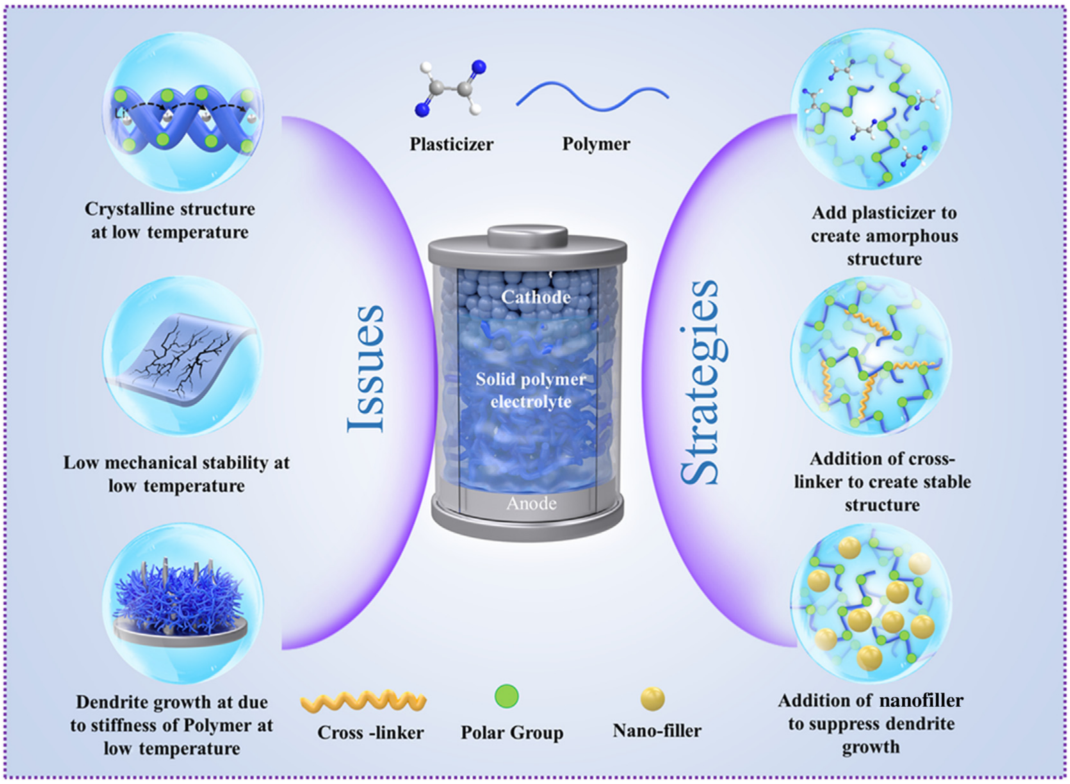

Figure 3 | *Limitations and strategies of inorganic SPE at low-temperature LBs.*

between lithium plating/stripping dynamics and slow ion conduction through the electrolyte, high-current dendrites can form locally. As a result of these dendrites penetrating the SEs, the battery may short-circuit.[47–51]

Several remedies have been developed to address this problem. It is possible to maintain uniform ion transport by tailoring the electrolyte materials with higher ionic conductivity at low temperatures, such as doped sulfide or ceramics-based electrolytes. Adding protective layers to the interface between lithium metal and the electrolyte can also be helpful in suppressing dendrite growth or inhibiting dendrite penetration.[52,53] It is also possible to prevent dendrite formation by applying external pressure to the lithium anode and electrolyte. A composite electrolyte merging both polymer and inorganic components offers a flexible, dendrite-resistant architecture that can enhance the accommodation of Li^+ movement and reduce dendritic growth.[54]

Furthermore, for SSLBs with inorganic SEs, interfacial contact remains a significant challenge. The polycrystalline cathodes contain pores because SEs do not have the fluidity and wettability of OLEs, so these gaps are not filled. During assembly and cycling, inorganic SEs may be too flexible, causing excessive pores and physical contact loss inside the solid composite electrode. There can be an adverse effect on electrochemical performance caused by limited interfacial contact in cold environments due to the drop in ionic conductivity as well as the possible variation of Young's modulus. The origin of the interfacial phenomenon in a solid system depends on two significant factors, the first of which is the mechanical robustness of the interface contact in a solid system, and the second of which is the chemical and electrochemical compatibility between the electrodes and SSEs.[55]

In one strategy to mitigate the interfacial resistance, we reduced the particle size of SE in the active materials to improve the solid/solid contact area between active materials (NCM712) and SE thus enhancing the Li^+ transport. In contrast to other configurations (L/S, L/L, and S/S), the configuration (S/L) coupling small-size (S) SE to the composite cathode and large-size (L) SE to the electrolyte layer exhibits superior electrochemical performance at room temperature and at −20 °C.[56] To further improve compatibility and to match the chemical and electrochemical properties of both the SSE and the electrodes, materials are often chosen or engineered in a manner to match both their chemical and electrochemical properties. A thin layer of a stable nitride or oxide can be applied to the electrode to act as a buffer layer, thus preventing direct contact and improving the interface between the electrode and the electrically conductive medium. Tailoring the configuration of the SSEs to certify chemical stability with the electrode and using additives or dopants to broaden the electrochemical stability window can also help to improve performance at low temperatures. These strategies are essential for maintaining high energy efficiency, reducing resistance, and extending the lifespan of SSBs in low-temperature environments.

SPEs hold substantial potential for low-temperature LBs because of their benefits over liquid electrolytes of inherent safety, light weight, and flexibility. The distinct chemical composition of polymer SEs and their resulting physicochemical properties distinguish them from their inorganic counterparts in terms of design strategies.[57,58] Some limitations and strategies to enhance the performance of SPEs are shown in Figure 3. Nevertheless, a great deal of ionic conductivity in SPEs is influenced by the mobility of polymer chains in subzero environments, which presents a challenge to their performance at low temperatures. As the temperature drops, the segmental motion of polymer chains is reduced. As a result, they become less flexible, which is vital for the conduction of ions in the polymer chains.[59,60] Generally speaking, in SPEs, ion transport is achieved by creating pathways for ions to hop or migrate through based on the movement of polymer chains. However, in a highly crystalline structure, the lack of free volume and mobility leads to lower ionic conductivity. For instance, SPEs are constructed on crystalline polymers, and the Li^+ transportation may arise within the noncrystalline patches of the polymers. Subsequently, sufficient ionic conductivity can be achieved in the subzero working environment by reducing the crystallinity of SPEs.[61] For example, incorporating LLZTO (Ta-doped $Li_7La_3Zr_2O_{12}$) fillers can decrease the crystallinity of the polymer matrix from 37.2% to 31.4%, while succinonitrile (SN) fillers can achieve a minimum of 8.4%.[62] The addition of the SN plasticizer will further boost the ionic conductivity of the system to 7.66×10^{-4} S cm^{-1}, which is more than four times higher than the effect that LLZTO fillers have on the ionic conductivity at room temperature. The plasticizer, however, softens the polymer matrix, thus reducing its mechanical strength. A softened structure at low temperatures can lead to a reduction in the material's ability to uphold its shape under stress when subjected to pressure and cycling, which is vital when it comes to applications such as SSEs, in which the electrolyte must endure pressure, mechanical deformation, and cycling stresses. As a result of the inferior mechanical strength of the battery, such issues are responsible for short circuits, cracking, and even electrolyte deformation. Hence, these limitations can be major factors that compromise the battery's overall stability as well as safety. To cope with the aforementioned challenges, Lv et al.[63] suggested that cross-linking could be used as an alternative strategy for enhancing mechanical strength and reducing crystallinity. However, with improved mechanical strength, crosslinking can also be responsible for potentially limiting ion transport by reducing polymer chain mobility. Optimal crosslinking needs to be applied to mitigate this problem, resulting in a strengthened polymer matrix without reducing ion mobility significantly. In this way, the electrolyte can maintain high ionic conductivity.

As far as the interfacial resistance is concerned, in spite of the fact that the ex-situ synthesized polymer SEs have high mechanical flexibility, one must still take into account the poor contact the polymer SEs have with other components of the cell due to this high interface resistance. To mitigate the aforementioned problem, Zhao et al.[64] used in-situ polymerization of 1,3-dioxolane to produce a conformal interfacial contact, which is initiated by cationic aluminum species. Good interfacial contact between the liquid precursors and electrode can be created by utilizing the wettability of these liquid precursors, which are then preserved after the polymerization process has been completed. In a recent study, Li et al.[65] confirmed that in-situ polymerized SEs with 1,3,5-trioxane-based precursors exhibit high capacity retention (more than 75% at −20 °C) and fast charge transfer kinetics. Notably, a reduction in the temperature affects the Li metal's mechanical properties, which impacts its interfacial contact with SE. In a brief but exciting study, Jolly et al.[66] investigated the deformability of Li metal at −20 °C, therefore necessitating a higher stacking pressure in order to obtain the same Li/SE interfacial contact at 25 °C, and hence, charge transfer resistance is decreased. Consequently, battery materials (such as SEs and Li metal anodes) and electrode/SE interface may become more crucial under SSB configuration in a low-temperature environment, which still requires further investigation.

Furthermore, another critical issue for SSLBs is dendrite growth in SPEs at low temperatures, which can compromise performance and battery safety. Dendrite penetration is primarily attributed to the soft nature of SPEs, which are also highly flexible and processable. Several factors contribute to dendrite growth, including growth at the tip or laterally, nucleation induced by subsurface structures, and redistribution of charges at the Li|SPE interface. Sharp dendrites quickly develop in the SPEs and eventually cause short-circuiting of the battery. A dendrite has a needle-like architecture that forms when lithium metals are charged and discharged repeatedly, particularly at low temperatures. SPEs with low temperatures tend to develop dendrites due to the redistribution of charge at the Li/SPE interface.[67] In a subzero environment, the nonuniform lithium ion transport in polymer electrolytes is attributed to the rigidity of the electrolyte and reduced ionic conductivity.[60] Consequently, lithium dendrite nucleation and growth occur due to localized high current densities caused by this uneven ion distribution. This issue is made more complicated by the low mobility of lithium ions within the stiffened polymer matrix, which makes dendrites more likely to penetrate the electrolyte and cause a short cycle.[68]

To address this challenge, several remedies have been explored. One methodology is to boost the ionic conductivity of the SPE at low temperatures by developing polymer composites or incorporating plasticizers that lead to more amorphous regions, allowing for better ion mobility. In addition, cross-linkers can be used to modify the polymer electrolyte structure, increasing mechanical strength without sacrificing flexibility and preventing dendrite penetration while maintaining mechanical strength.

In order to enhance the mechanical strength and ionic conductivity of SPE, various additives are commonly composited with it. This was the case not only with additives, also known as fillers, which were limited to ion-conductive materials. The morphology and size ranged from micro to nanometer and from three-dimensional, two-dimensional, one-dimensional, to zero-dimensional. Some properties of the fillers, such as thermal stability and chemical and electron insulation, are highly desired. Even if several techniques have been used recently to increase SEs' low-temperature ionic conductivity, collective strategy methods are still needed to achieve superionic conductivity and integrate techniques across numerous scales to collect the

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Table 1 | *Summary of the Critical Challenges and Effective Strategies of SSEs for Low-Temperature Li-Ion Batteries*[69–71]

Types	Polymer Solid Electrolytes	Inorganic Solid Electrolyte	Composite Solid Electrolyte
Shortcomings	1. The mobility of polymer chains at low temperatures 2. Dendrite growth in SPEs at low temperatures 3. Reduced mechanical strength is attributed to the softening of the polymer matrix 4. Poor contact with polymer SEs have with other components	1. Grain boundaries that result from their polycrystalline nature 2. The mobility of ions and atoms at low temperatures is significantly reduced, which causes grain boundaries at low temperatures 3. The growth of Li dendrite at low temperature 4. Interfacial contact between the electrolyte and electrode	1. CSEs exhibit reduced ionic mobility at low temperatures, leading to lower overall conductivity. 2. Inorganic fillers may undergo degradation at the interfaces, particularly at low-temperatures 3. The differences in thermal expansion coefficients of polymer and inorganic components may lead to microcracks during repeated temperature cycling
Strategies	1. Incorporating fillers and plasticizer can decrease the crystallinity of the polymer matrix 2. The addition of an optimized cross-linker can increase the mechanical stability as well as ionic conductivity 3. In-situ polymerization is attributed to producing a conformal interfacial contact initiated by cationic aluminum species. 4. The addition of conductive filler can suppress dendrite growth.	1. Doping SEs with foreign cations 2. Partial substitution of anion composition 3. Coating or modifying grain boundaries with highly conductive secondary phases 4. Adding protective layers to the interface. 5. Reducing the particle size of SE in the active materials	1. Use nano-sized fillers with high ionic conductivity to enhance ion transport. 2. Incorporate polymers with low glass transition temperature 3. Polymers (e.g., PEO blends or ionic polymers) to maintain flexibility at low temperatures.
Representative materials	PEO based electrolytes	LSiGePSBrO	PEO-based CSE

lowest activation energy. Li^+ carriers' interactions with the anion framework or surrounding coordination configuration, the impact of grain boundaries or other defects on Li^+ transport, and the lessons learned from developing low-temperature OLEs (such as desolvation energy and the comprehension of solvation structure) should all be carefully considered, particularly with regard to the low-temperature atmosphere (Table 1).

Summary and Outlook

In summary, the research evolution of SSEs in subzero environments with respect to kinetic processes, fundamental requirements, critical challenges, and effective material and chemistry design strategies has been methodically reviewed in this perspective . Basically, Li^+ migration in SE may be divided into two categories based on the kinds of SE: bulk and grain boundary transport. Further, the interfacial kinetic process at the SE/electrode interface is governed by three main steps: Li^+ diffusion across the interphase, Li^+ transport across the SE/interphase interface, and charge transfer at the electrode surface. These steps are closely interconnected due to the complexity of the SE/electrode interface. One way to describe the limitations with low-temperature SSBs is as follows: (1) slow solid-state Li^+ diffusion in electrode materials; (2) limited interfacial contact and exacerbated contact loss problems; (3) high interfacial resistance resulting from an unstable interface or unfavorable interphases; and (4) the sharp decline in SE's ionic conductivity as the temperature drops.

To overcome the aforementioned limitations, several tactics has been developed regarding SEs, such as tailoring the materials and chemistry design. In addition to increasing room-temperature ionic conductivity, grain boundary resistance of SEs and lowering the activation energy is advantageous for preserving high ionic conductivity at low temperatures. It has been suggested that the use of additives or doping, interphase modification, additional cross-linker, plasticizer, nanofiller, and enhancement of interfacial contact can improve the interfacial kinetics at low temperatures at the SE/electrode interface. The following promising pathways are suggested in light of the comprehensive knowledge of kinetic processes, low-temperature constraints, and design methods of the SEs of LBs (Table 2):

1. To ascertain the effective working temperature constructed in the unified charge/discharge procedures, a low-

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Table 2 | *Summary of the Electrochemical Performances of Solid-State Electrolyte Operating at Low Temperature*

Electrolyte	Ionic Conductivity (mS cm^{-1})	Activation Energy (eV)	Specific Capacity (mAh g^{-1})	Temperature (°C)	Ref
Poly(vinyl ethylene carbonate) polymer electrolyte	0.4	0.159	104	−15	[72]
1.5Li_2O-$HfCl_4$	0.34	0.275	140 at 1C	−10	[73]
PP6LS20@GF	0.17	0.115	97.2	−20	[74]
Li_3InCl_6	0.1	0.326		−25	[75]
S-LHCE	0.031	—	115 at 0.1C	−10	[76]
HT150-5PMMA	0.01	0.48		−35	[77]
Li_6PS_5Cl	0.6	0.30		−20	[78]
$Li_{9.54}[Si_{0.6}Ge_{0.4}]_{1.74}P_{1.44}S_{11.1}Br_{0.3}O_{0.6}$	1.9	0.24	17.3 mAh cm^{-2}	−10	[79]
LASI-80Si	3.9	0.2	700	0	[80]
Ceramic polymer composite electrolytes	0.05	0.21	154.5 at 0.1C	−20	[81]
TA-star-PEGMEMA/LiAMPS	0.19×10^{-2}	28.06	145 at 0.1 C	0	[82]
Crystalline $Li_{5.5}PS_{4.5}C_{l1.5}$	0.51	0.22	104.8^1 at 0.02 C	−20	[83]
Li_3InCl_6	0.16	0.34	77.6	−10	[84]
Li_6PS_5Cl	0.13	0.395	110.5	−30	[85]
L-SE($Li_{5.5}PS_{4.5}Cl_{1.5}$)	0.52	0.25	89 at 0.05 C	−20	[56]

temperature performance appraisal standard should be recognized for the critical indicators (e.g., capacity retention ≥80% and ≥50% at −40 °C and −60 °C, respectively).

2. Establish a precise and logical impedance analysis technique to measure the activation energy of each kinetic process and separate the interfacial responses from the interphase layer and charge transfer process from the anode/SE and cathode/SE interface.
3. Improve low-temperature performance, advanced SEs with low activation energy and high ionic conductivity must be developed.
4. Elucidate the interphase development at low temperature and the true microscopic kinetic mechanism at the interface.
5. Investigate the chemo-mechanical limitations at subzero environment, and accordingly develop tactics to resolve/improve the interfacial contact issues.

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