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Challenges and Progress of Solid-State Electrolytes for Silicon-Based Solid-State Batteries

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Abstract. Silicon-based solid-state batteries provide an attractive route for developing lithium batteries with higher energy density and better safety, but their practical progress is still limited by the difficult match between silicon anodes and solid-state electrolytes. The central issue is not only the intrinsic volume change of silicon, but also the unstable Si/SSE interface produced during repeated lithiation and delithiation. Once silicon expands and contracts, the solid electrolyte cannot wet or refill the interface like a liquid electrolyte, which easily leads to contact loss, void formation, interrupted Li+ and electron transport, impedance growth, and rapid capacity fading. This review therefore focuses on oxide, sulfide, and polymer/composite electrolytes coupled with silicon-based anodes. Oxide electrolytes generally possess good chemical stability and wide electrochemical stability windows, but their rigid and brittle nature makes it difficult to maintain continuous contact with the deformable silicon electrode. Sulfide electrolytes show high ionic conductivity and better mechanical deformability, especially under stack pressure, yet their air sensitivity, interfacial reactions with silicon, carbon-induced decomposition, and pressure-dependent performance remain serious obstacles. Polymer and polymer-based composite electrolytes are more flexible and easier to process, which gives them certain advantages in accommodating silicon volume variation, but their low room-temperature ionic conductivity, limited mechanical strength, and long-term interfacial degradation still need to be addressed. In this view, the development of silicon-based solid-state batteries cannot rely on improving one component alone. More attention should be given to the coordinated design of electrolyte composition, interfacial chemistry, and electrode architecture, so that a stable, low-resistance, and mechanically adaptive Si/SSE interface can be achieved for practical high-energy solid-state batteries.

Keywords: Silicon-based anodes, Solid-state batteries, Solid-state electrolytes, Si/SSE interface, Interfacial engineering

1. Introduction

The rapid expansion of portable electronics, electric vehicles, and grid-scale energy storage has placed increasing demands on lithium-ion batteries in terms of energy density, safety, cycle life, and cost. Commercial lithium-ion batteries based on graphite anodes and organic liquid electrolytes have achieved great success [1], but this configuration is approaching its practical limits. The limited capacity of graphite has gradually become a clear constraint on further increasing the energy density of lithium-ion batteries. At the same time, the use of liquid electrolytes still leaves a safety concern that cannot be ignored, since organic solvents may leak, evaporate, burn, or even trigger thermal runaway under harsh operating conditions [2, 3]. From this point of view, solid-state electrolytes are more than a simple replacement for liquid electrolytes. They offer a practical route to improve cell safety while also leaving room for the design of batteries with higher energy density [4, 5].

The anode largely sets the upper limit of battery energy density, so its selection is especially important in the design of high-energy solid-state batteries. Lithium metal appears very attractive in this regard, mainly because it delivers an ultrahigh theoretical capacity and has an extremely low electrochemical potential. In practice, however, it is still not easy to make lithium metal work reliably in solid-state cells. Dendrite growth, continuous interfacial side reactions, dead lithium accumulation, and poor physical contact with solid electrolytes can gradually damage the Li/SSE interface and raise safety concerns [6, 7]. Silicon-based anodes therefore provide a more practical alternative in some cases. They retain a high theoretical capacity while also offering a relatively low lithiation/delithiation potential, abundant reserves, and better environmental compatibility. These features make silicon-based anodes promising for high-energy-density solid-state batteries, provided that their volume change and interfacial stability can be properly managed [8]. In addition, the working potential of silicon is higher than the lithium plating potential, which can reduce the risk of dendrite-induced short circuits compared with lithium metal anodes.

Despite these advantages, silicon anodes introduce their own set of challenges. In liquid-electrolyte batteries, silicon suffers from repeated solid electrolyte interphase (SEI) formation, particle pulverization, loss of electrical contact, and poor rate capability caused by large volume variation, low electronic conductivity, and slow Li^+ diffusion [9]. In solid-state batteries, the problems of silicon anodes are mainly concentrated at the solid–solid interface. Unlike liquid electrolytes, which can rewet newly exposed surfaces and pores during cycling, solid-state electrolytes rely largely on the physical contact formed during cell assembly. This limitation becomes especially severe for silicon, since its repeated expansion and contraction can open voids and gaps at the Si/SSE interface. Once the original contact is weakened, Li^+ migration across the interface becomes discontinuous, and the electronic connection within the silicon electrode may also be disturbed. As a result, interfacial resistance increases and capacity decay is accelerated [10]. Thus, the silicon anode itself explains only part of the performance of silicon-based solid-state batteries. Electrolyte chemistry, mechanical adaptability, and the ability to preserve Si/SSE contact during long-term cycling are just as important.

This review therefore discusses oxide, sulfide, and polymer/composite solid-state electrolytes from the viewpoint of their compatibility with silicon anodes. Instead of simply comparing them as separate electrolyte categories, it focuses on how each system behaves at the Si/SSE interface during operation, especially in terms of interfacial stability, mechanical matching, and Li^+ transport continuity. The discussion then clarifies the main limitations of each electrolyte system and points out interface-design strategies that may be more realistic for long-term silicon-based solid-state batteries.

2. Oxide solid-state electrolytes for silicon-based solid-state batteries

2.1. General features of oxide solid-state electrolytes

Oxide solid-state electrolytes are among the earliest inorganic electrolyte systems investigated for solid-state batteries. Typical examples include LiPON-type electrolytes, NASICON-type

LATP/LAGP, perovskite-type LLTO, and garnet-type LLZO/LLZTO. Their main strength lies in their chemical robustness. Compared with many sulfide and polymer electrolytes, oxide electrolytes usually show better air stability, thermal stability, and wider electrochemical stability windows, which makes them attractive from a safety perspective. LATP and LAGP can reach room-temperature ionic conductivities on the order of 10^{-4} S cm^{-1}, while LLZO has also received broad attention because of its relatively high Li^+ conductivity and wide electrochemical stability window [11, 12].

The ceramic nature of oxide electrolytes brings both benefits and drawbacks. On the positive side, it gives these materials good chemical stability and a relatively wide electrochemical stability window. On the other hand, the same rigid structure makes them difficult to pair with silicon anodes. Most oxide electrolytes are hard and brittle, so they usually cannot build intimate contact with composite electrodes through simple pressing or conventional processing. This mismatch becomes more pronounced during cycling. Silicon expands and contracts repeatedly, while the oxide electrolyte has little ability to follow this deformation. As a result, the originally formed Si/SSE contact may gradually break down, leaving local gaps, contact-loss regions, and high-resistance interfaces. Once this happens, Li^+ transport across the solid–solid interface is strongly hindered, even when the oxide electrolyte itself has acceptable bulk ionic conductivity [13].

2.2. Challenges with silicon-based anodes

For oxide-based silicon solid-state batteries, the most difficult problem is the mechanical mismatch at the Si/SSE interface. In conventional liquid-electrolyte cells, fresh surfaces, cracks, or pores formed during cycling can still be wetted by the electrolyte to some extent. Oxide electrolytes do not have this self-adjusting ability. Once silicon expansion creates voids or weakens the original contact, the oxide electrolyte cannot flow into these newly formed spaces or rebuild the interface. As a result, the Si/SSE contact in oxide-based systems is often limited to the initially formed contact area. It behaves more like a constrained two-dimensional solid–solid interface, rather than a fully infiltrated three-dimensional contact network [14].

Because silicon undergoes a volume change of more than 300% during lithiation/delithiation, this rigid interface is highly vulnerable to cycling-induced damage. Silicon expansion can generate local stress at the interface, while silicon contraction can leave interfacial voids. These processes lead to contact loss, stress concentration, increased interfacial impedance, and blocked Li^+ transport. Therefore, improving oxide-based silicon solid-state batteries requires more than increasing the intrinsic ionic conductivity of the electrolyte. A stable oxide/Si interface must also accommodate volume variation, maintain contact, and suppress interfacial resistance growth.

2.3. Research progress of different oxide solid-state electrolytes

Different oxide electrolyte systems have been explored to improve the compatibility between silicon anodes and solid electrolytes. LiPON is one of the most representative oxide-type electrolytes for thin-film solid-state batteries. Ke et al. prepared an approximately 3 μm-thick LiPON film by radio-frequency reactive magnetron sputtering and integrated it with a 70 nm Si thin-film anode. The cell retained 74.5% of its capacity after 100 cycles [15]. Such a compact layer can passivate the Si surface and reduce direct side reactions with the electrolyte. However, this advantage is mostly limited to thin-film cells. LiPON has low room-temperature ionic conductivity and usually requires vacuum deposition, which is costly, slow, energy-intensive, and difficult to scale up [16]. Because of these limitations, LiPON is usually more practical as a thin-film electrolyte or artificial interfacial layer. Its use in high-loading thick electrodes remains difficult.

Among oxide electrolytes, garnet-type LLZO and LLZTO are often considered more promising for high-energy cells because their bulk ionic conductivity and electrochemical stability [17]. Ferraresi et al. reported a Si/LLZTO cell with a high initial capacity, but the capacity decreased significantly after 100 cycles [18]. This result shows that a well-defined initial interface, even one produced by thin-film deposition, may still fail once silicon repeatedly changes volume. For ultrathin Si layers, the interfacial strain is relatively limited, and stable contact can be maintained to some extent. When silicon loading or electrode thickness increases, however, repeated expansion and contraction can quickly damage the Si/SSE interface. For LLZO/LLZTO-based systems, forming a clean starting interface is only the first step; the harder task is to keep that interface intact during long-term cycling.

NASICON-type LATP and LAGP offer a different oxide-electrolyte route because of their relatively high ionic conductivity, lower cost, and good air stability. Their direct contact with silicon, however, introduces another problem. At the Si/LATP interface, interdiffusion and alloying reactions can occur, leading to fast interfacial deterioration and impedance growth [19]. Liu et al. addressed this issue by coating the LATP surface with graphene. The graphene layer worked as a barrier against Si diffusion into LATP, thereby improving interfacial stability [20]. This case suggests that LATP/LAGP may be more useful as interfacial or composite components than as dense ceramic electrolytes in direct contact with silicon.

2.4. Summary of oxide electrolytes

For silicon-based solid-state batteries, oxide electrolytes present a clear trade-off. Their chemical, thermal, air, and electrochemical stability are valuable, but their rigid ceramic nature is poorly matched with a silicon anode that repeatedly expands and contracts. LiPON can build stable thin-film interfaces, but its low conductivity and costly deposition limit its use in thick electrodes. LLZO/LLZTO provide better intrinsic conductivity and stability, yet their Si/SSE interfaces are still difficult to preserve under repeated silicon volume change. LATP/LAGP are attractive in terms of cost and air stability, but direct Si contact may trigger interdiffusion and alloying reactions. Therefore, future oxide-based designs should focus less on the oxide electrolyte alone and more on flexible buffer layers, artificial interfacial coatings, and composite structures that can maintain contact while reducing interfacial resistance.

3. Sulfide solid-state electrolytes for silicon-based solid-state batteries

3.1. General features of sulfide solid-state electrolytes

Sulfide solid-state electrolytes have become highly attractive for silicon-based solid-state batteries because they combine high room-temperature ionic conductivity with better mechanical deformability than oxide electrolytes. Sulfide electrolytes are generally softer and can undergo partial plastic deformation under external pressure. This feature allows them to fill interparticle voids and improve solid–solid contact inside composite electrodes. As a result, sulfide electrolytes can partly reduce the rigid-contact failure that limits oxide-based systems and are widely considered promising electrolytes for Si-based solid-state batteries.

3.2. Main challenges in Si/sulfide systems

Although sulfide electrolytes offer favorable transport and contact properties, their integration with silicon anodes is still challenging. One issue comes from electrode composition. Silicon has limited intrinsic electronic conductivity and slow Li^+ diffusion, so thick silicon electrodes usually require both electronic conductive additives and sulfide electrolyte particles to build continuous electron/ion transport pathways. However, most of these additives do not contribute directly to electrochemical capacity. If their content is too high, they may dilute the active-material fraction in the electrode and lower the practical energy density of the full cell.

The Si/sulfide interface is another weak point that needs careful attention. During cycling, the repeated expansion and contraction of silicon can gradually loosen the interfacial contact. At the same time, sulfide electrolytes may react with silicon or with other electrode components, forming interphase layers with poor ion transport. These newly formed layers are not always beneficial. Once they become thick or uneven, they can raise interfacial impedance, reduce Coulombic efficiency, and accelerate capacity decay during long-term cycling [21, 22]. Carbon additives are usually introduced to improve electronic conduction in silicon-based sulfide systems, especially when the silicon particles are poorly connected to each other. But carbon isn't always beneficial in sulfide environments—it can speed up electrolyte decomposition and complicate interfaces. That creates a tricky balance: the electrode needs good conduction for both Li^+ and electrons, but without triggering excessive side reactions or sacrificing active silicon. So designing sulfide-based silicon solid-state batteries really means juggling transport efficiency, interfacial stability, and practical silicon loading.

3.3. Research progress in sulfide solid-state electrolytes

Recent efforts to boost sulfide-based silicon solid-state batteries have zeroed in on redesigning the electrode recipe. The old approach—just tossing together silicon, carbon, and sulfide electrolyte—is giving way to smarter multicomponent systems where one ingredient pulls multiple duties. Ideally, that ingredient should shuttle Li^+, keep electrons connected, and also take the edge off silicon's notorious swelling during cycling. Dunlap et al. tried heattreated polyacrylonitrile (PAN) as a mixed ionicelectronic conductor in a silicon anode. In their setup, PAN wasn't merely a binder holding things together; it actually wove itself into the transport network, making it easier for charges to move through the electrode [23]. Yan et al. found that adding hard carbon helps stabilize Li–Si alloy anodes. The pores and graphitic patches inside hard carbon soak up excess lithium and keep dendrites—or at least soft shorts—in check. As a result, their NCM811/Li_6PS_5Cl/Si–HC full cells ran stably for many more cycles than without carbon [24]. These examples show a real shift: no more just mixing Si, C, and electrolyte. Instead, each component in the electrode handles one job—transport, stability, or buffering.

Interfacial engineering is another important direction. The challenge is to limit direct Si/sulfide contact and suppress side reactions without sacrificing Li^+ transport. Chen et al. inserted a SiS_2modified $Li_7P_3S_{11}$ layer between the silicon electrode and the bulk sulfide electrolyte [25]. This design echoes a broader trend in sulfidebased silicon anodes: moving from simple physical mixtures toward integrated composites, where each component contributes to transport, stability, or mechanical buffering.

Other studies have tried to reduce the amount of sulfide electrolyte directly mixed into the silicon anode. Xu et al. reported a nano-Si anode for sulfide-based all-solid-state lithium-ion batteries without adding extra electrolyte into the anode [26]. This work shows that stable transport does not necessarily require a high electrolyte fraction in the electrode. By controlling the Si/electrolyte boundary and using a nanoscale Si architecture, the electrode can maintain ion and electron pathways while retaining a higher active-silicon content. For Si/sulfide systems, this suggests a more practical direction: the interface should be kept thin, stable, and sufficiently Li^+-conductive, rather than relying on excessive inactive electrolyte to compensate for poor contact.

3.4. Summary of sulfide electrolytes

Sulfides beat oxides in ionic conductivity, compressibility, and solid–solid interfacial adaptability—traits that make them attractive for silicon-based solid-state batteries. Yet moisture sensitivity, interfacial side reactions, carbon-induced decomposition, void formation during cycling, and the need for external stack pressure still hold them back.

Future works should focus on theses aspects. One is keeping Si/sulfide contact stable even with little or no external pressure. Another is building efficient Li^+/electron networks without relying on excess inactive carbon or electrolyte. A third: taming interfacial chemistry to suppress sulfide decomposition and make the materials processable in air. Tackle these, and sulfide-based silicon solid-state batteries can realistically hit high-energy targets—not just stay in the lab.

4. Polymer/composite solid-state electrolytes for silicon-based solid-state batteries

4.1. General features of polymer/composite solid-state electrolytes

Compared with ceramic electrolytes, polymer-based electrolytes are attractive for silicon anodes mainly because they can form more compliant contacts with dynamically changing electrode surfaces. Their low density, film-forming ability, processability, and interfacial wettability make them easier to integrate with silicon electrodes. More importantly, their elastic and plastic deformation can help accommodate silicon expansion and contraction, thereby delaying interfacial debonding during cycling [27, 28].

For silicon-based anodes, this mechanical compliance is particularly valuable. Polymer electrolytes can avoid the rigid–rigid contact typical of oxide-based systems and may maintain stable contact under low or even no external stack pressure. Therefore, polymer and organic–inorganic composite electrolytes are important candidates for improving Si/SSE interfacial compatibility.

4.2. Main challenges of polymer electrolytes

The main limitation of polymer electrolytes is that their interfacial flexibility is often obtained at the cost of weaker ion transport and mechanical strength. Li^+ migration in polymers mainly depends on polymer-chain segmental motion, which is limited at room temperature. As a result, polymer electrolytes usually show much lower room-temperature ionic conductivity than liquid electrolytes and many inorganic solid electrolytes. Their typical room-temperature conductivity is reported to be only 10^{-8}–10^{-5} S cm^{-1} [29].

In addition, some polymer electrolytes have narrow electrochemical stability windows, making it difficult to pair them simultaneously with high-voltage cathodes and low-potential silicon anodes. Their long-term chemical and mechanical stability is also insufficient in some cases. During repeated cycling, silicon volume changes can continuously impose tensile, compressive, and shear stresses on the polymer interface. Therefore, polymer electrolytes face a conductivity–flexibility trade-off: they provide better interfacial compliance than ceramic electrolytes but still need higher bulk conductivity, stronger mechanical support, and improved long-term durability [30].

4.3. Research progress of polymer/composite solid-state electrolytes

To overcome these limitations, researchers have developed several strategies to improve Li^+ transport, interfacial contact, and mechanical adaptability. Recent polymer-based strategies have therefore moved beyond simple conductivity enhancement. Approaches such as gel component incorporation, in situ polymerization, copolymer regulation, inorganic filler addition, and self-healing network construction are now being used to improve both Li^+ transport and interfacial durability. In the case of silicon anodes, this shift is particularly important, because a polymer electrolyte must not only conduct ions efficiently, but also remain in close contact with the deforming Si surface during repeated cycling.

A useful way to improve silicon-based solid-state batteries is to create continuous Li^+ transport pathways within the electrode itself, rather than relying only on ion conduction through the bulk polymer electrolyte. Dong et al. used micron-sized porous Si/C electrodes together with a PEO-LiTFSI electrolyte. After prelithiation and pre-infiltration, the electrolyte could penetrate more effectively into the porous electrode, forming internal Li^+ transport channels and leading to improved electrochemical performance [31]. In another study, Zhang et al. prepared Si@VG anodes by growing vertical graphene sheets on silicon nanoparticles and then paired them with PEO-based solid electrolytes. The vertical graphene network helped maintain electronic connection between silicon particles and, at the same time, improved the physical contact between the electrode and electrolyte [32]. These examples show that polymer electrolyte optimization should not be treated separately from electrode design. For silicon anodes, a more convincing strategy is to build an integrated ion/electron transport network while preserving stable electrode/electrolyte contact during cycling.

In situ polymerization and gel polymer electrolytes offer a more contact-friendly strategy for silicon-based solid-state batteries. Unlike preformed polymer membranes, which may only contact the outer surface of the electrode, in situ polymerization allows the electrolyte precursor to penetrate into the electrode pores before solidifying. This process helps build closer electrode/electrolyte contact, reduces interfacial gaps, and provides more continuous Li^+ transport pathways. Wang et al. used $LiNO_3$ to regulate the ring-opening polymerization of DOL, leading to the formation of a two-dimensional SEI on the silicon surface together with a high-concentration polymer electrolyte [33]. In this design, the improved reversibility of silicon mainly comes from a more stable interfacial chemistry rather than from polymer conductivity alone. Qiu et al. further combined double-layer carbon coating, chemical prelithiation, and DOL in situ polymerization to construct Si@C@C anodes coupled with PDOL polymer electrolytes. This integrated strategy improved both cycling stability and Coulombic efficiency by simultaneously stabilizing the silicon surface and strengthening electrode/electrolyte contact [34].

Mechanical adaptability has also become an important consideration in polymer electrolyte design, especially for silicon anodes that undergo repeated volume changes. He et al. reported a self-healing dynamic supramolecular elastomer electrolyte, which could also work as a binder in silicon-based electrodes [35]. Zhao et al. constructed a quasi-solid-state electrolyte through LiDFOB-induced ring-opening polymerization of 1,3,5-trioxane. This electrolyte achieved an ionic conductivity of 2.16×10^{-3} S cm^{-1} and a Li^+ transference number of 0.61. At the same time, LiDFOB-derived SEI formation suppressed side reactions and reduced volume expansion of the Si/C anode after cycling [36].

Overall, current polymer/composite electrolyte research has shifted from single-property optimization to integrated interface design. In situ polymerization, gel networks, inorganic fillers, self-healing structures, and stable SEI chemistry are all being used to build more adaptive Si/SSE interfaces.

4.4. Summary of polymer/composite electrolytes

Polymer and composite electrolytes show clear advantages in flexibility, processability, and stress-buffering capability. These properties can alleviate the rigid-contact issue of oxide electrolytes and improve interface retention during silicon volume change. However, polymer-based systems still face low room-temperature ionic conductivity, limited electrochemical stability, insufficient mechanical strength, long-term interfacial instability, and filler-dispersion issues in composite electrolytes.

Future polymer/composite electrolyte design should balance high ionic conductivity, strong mechanical support, interfacial flexibility, and low interfacial resistance. Room-temperature operation, thick-electrode compatibility, and self-adaptive interfacial repair should be emphasized for practical silicon-based solid-state batteries.

5. Conclusion

Silicon-based solid-state batteries are attractive because they combine the high capacity of silicon anodes with the safety advantage of solid-state electrolytes. In real cells, however, this combination does not necessarily lead to stable performance. Silicon undergoes repeated expansion and contraction during cycling, and this dimensional change can gradually damage the Si/SSE interface. Once local contact is lost, Li^+ migration across the interface becomes less efficient, while the electronic connection inside the electrode may also be disturbed. These effects usually appear together and finally lead to higher interfacial resistance and faster capacity decay. Therefore, improving only the electrolyte conductivity or choosing a chemically stable electrolyte is not sufficient. In my view, the key point is to design the electrolyte chemistry, interfacial structure, and electrode architecture as a whole, so that ion transport, mechanical contact, and interfacial stability can be maintained at the same time. Only when ion transport, mechanical contact, and interfacial stability are considered at the same time can the capacity advantage of silicon be more effectively translated into durable solid-state battery performance.

For oxide-based systems, the next step should not be limited to pursuing higher bulk ionic conductivity. More attention needs to be given to flexible buffer layers and lower-cost composite designs, because these strategies are more likely to relieve the mechanical mismatch between rigid oxides and expanding silicon. In sulfide-based systems, the key issues are somewhat different. Their high ionic conductivity is already attractive, but air sensitivity and interfacial chemical reactions with silicon still need to be better controlled. For polymer and polymer-based composite electrolytes, the main task is to raise room-temperature ionic conductivity without sacrificing mechanical durability during long-term cycling. From my perspective, the practical use of silicon-based solid-state batteries will depend on whether these different design factors can be balanced in real cell configurations. Material properties, interfacial stability, electrode structure, and scalable processing should be considered together, rather than optimized separately. Only in this way can the high-capacity advantage of silicon be translated into solid-state batteries with high energy density, improved safety, and realistic manufacturing potential.

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