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Interfacial Engineering Strategies for Sulfide-Based All-Solid-State Lithium Batteries: From Particle Design to Surface Modification

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Abstract. With the rise of electric vehicles, safer and high-energy-density energy storage solutions have become increasingly important and urgent. Under this trend, research on sulfide-based all-solid-state lithium batteries (ASSLBs) has been continuously deepened to replace traditional lithium-ion batteries. In sulfide-based ASSLBs, sulfide solid electrolytes (SSEs) have excellent mechanical compliance and high ionic conductivity, but they also face challenges such as mechanical degradation, contact loss, and interface instability. This paper examines three critical new developments, including interface control during fabrication level, surface coatings and artificial interface layers, and joint particle and microstructure design. Case histories reveal ways in which bimodal particle morphology, oxide-related coatings, and solvent-free dry electrode manufacturing raise conductivity, strength, and manufacturability. But these technologies continue to have limitations such as inability to simultaneously balance particle size and reactivity, issues with scale and uniformity of coatings, and limitations of most dry processing technologies in mechanical and environmental conditions. Therefore, to counter these limitations, an overall interface engineering solution involving dry processing, multi-functional coatings, and particle optimization is proposed in this paper to extract synergistic benefits, improved stability, and scalability. This solution provided an achievable path towards sulfide-based ASSLBs with longer cycle life, improved energy density, and commercial viability.

Keywords: Sulfide Electrolytes, All-solid-state Lithium Batteries, Interfacial Engineering

1. Introduction

All-solid-state lithium batteries (ASSLBs) are regarded as the best alternative to traditional lithium-ion batteries (LIBs) due to their enhanced safety, widened electrochemical window, and high energy density [1]. Within ASSLBs, sulfide-based solid-state electrolytes (SSEs), such as Li_6PS_5Cl, exhibit enhanced ionic conductivity and superior mechanical compliance with respect to oxide or polymer electrolytes [2]. For these reasons, they are regarded as the best possible candidates for viable ASSLBs. However, there are also a number of issues remaining in the electrode-electrolyte interface in sulfide-based ASSLBs, such as chemical instability, interface contact loss, and mechanical decay during the course of electrochemical cycles, all of which must be addressed promptly [3]. For the

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proper solution of the above issues and the realization of the best possible potential of sulfide-based ASSLBs, interface engineering has been adopted as the basic rule of design in most researches so far. This review also presents the latest information in the above four research branches with respect to interface engineering: interface failure mechanisms, particle and microstructure designs, artificial buffer layers, and optimization in the fabrication level.

Between positive electrode material such as high-nickel layered oxides and sulfide electrolytes (SSE), the interface layer tends to lose stability, which has been a general problem in sulfide-based ASSLBs. Interface reactions tend to yield resistive products, raising battery impedance rapidly and leading to capacity fade [4]. Moreover, formation of deformation during cycle operation tends to cause the sulfide electrolyte, which is prone to brittleness, to crack and thus battery failure [5]. Further, sulfide electrolytes also exhibit other means of instability which were not addressed above. Having an all-encompassing solution capable of withstanding mechanical strain and supporting simultaneous electrochemical reactions becomes paramount due to prevalent mechanical failure and chemical decomposition in batteries.

Particle and microstructure engineering provides an effective means to customize the mechanical strength and electrochemical property of all-solid-state lithium batteries (ASSLBs). Schlautmann et al. [6] demonstrated size reduction of solid electrolyte (SE) particles to eliminate pore spacings between particles and increase the ionic conductivity with better interface contact. Hong et al. [7] also proposed a bimodal cathode structure composition with polycrystalline particles and single-crystal particles to reduce microcracks and increase the packing density. The study displays promising potential to reinforce charge transport, mechanical integrity, and long-term cycle life with better morphology and particle size distribution in composite electrodes.

Some studies have determined that an artificial interface layer and surface coating are an effective measure of retaining surface reactivity in sulfide-based all-solid-state batteries. Liang et al. [3] have suggested that oxide coatings like $LiNbO_3$ and Al_2O_3 increase ionic transport and inhibit interfacial decay [8]. In contrast, Hood et al. [9] have suggested that new developments in atomic layer deposition (ALD) make conformal coatings of Al_2O_3 over Li_6PS_5Cl powder feasible, increasing ionic conductivity through doubling but stopping moisture decay [9]. Therefore, ultra-thin conformal coatings have great potential in interface control. However, these technologies are difficult to achieve the final effect of scalable, uniform deposition without sacrificing mechanical durability.

Moreover, process-level techniques like dry electrode fabrication have shown promise in improving sulfide-based ASSLBs' manufacturability and interfacial contact. Dry processing makes it possible to create dense, solvent-free electrode designs with improved ionic channels and mechanical stability in contrast to slurry-based techniques. Maintaining the integrity of moisture-sensitive sulfide electrolytes such as Li_6PS_5Cl while operating in ultra-dry circumstances (≈ -40 °C dew point) makes them compatible with roll-to-roll manufacturing. Though there are still issues with humidity management, binder selection, and scaling to large sizes without causing microstructural damage or loss of cohesiveness, methods like hot rolling and calendaring can maximize electrode uniformity and thickness, as recent studies have described [10].

While surface coatings, process-level controls, and particle/microstructure co-design have all improved sulfide-based ASSLBs, significant issues still exist. Particle size, reactivity, and scalability are trade-offs in particle engineering, and consistent SC/PC ratios remain difficult to achieve. Because wet techniques lack homogeneity and ALD is still expensive, coating schemes that suppress interfacial reactions struggle with thickness control, cracking, and scalability. Although technique necessitates ultra-dry rooms, precise binder adjustment, and pressure control, dry electrode

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production increases manufacturability. For ASSLBs to be scalable, long-lasting, and economically viable, these problems must be fixed.

This paper suggested a comprehensive interfacial engineering framework that incorporates tactics from several scales, ranging from surface modification (chemical coatings, artificial interphases) to particle design (size, morphology, distribution), in order to overcome these constraints. This research will map out the ways in which these methods impact both ionic transport and mechanical integrity at the interface. In addition to increasing cycle life and energy density, such an integrated approach has the potential to direct the scalable production of next-generation solid-state batteries for practical use.

2. Interfacial engineering strategies in sulfide-based ASSLBs

One fundamental tactic to deal with interfacial instability and mechanical deterioration in ASSLBs is the tailoring of particle shape and microstructure. The optimization of particle size and distribution is a primary concern since it has a direct impact on the composite electrodes' mechanical integrity, interfacial contact, and electrochemical performance. Schlautmann et al. [6] demonstrated that reducing the particle size of the solid electrolyte (SE) and the cathode active material (CAM) can enhance the diffusion effect and reduce the tortuosity, thereby increasing the interface contact area, broadening the Li^+ transport path and reducing the interface resistance. But excessively reducing particle size also leads to an excessively increased surface reactivity, causing counterproductive side effects such as reducing the lifespan of the battery. Comparatively, Hong et al. [7] constructed a bimodal particle engineering technique, which is a more systematic and refined technique. This technology customizes the composition of large polycrystalline (PC) particles and tiny single-crystalline (SC) particles to assemble high-density electrodes with high mechanical strength. In Hong et al.'s research, the bimodal cathode composed of nickel-rich layered oxide (NCMA) active material exhibited notable dominance over the unimodal cathode. This technique of plugging tiny SC particles (D_{50} = 4.58 μm) with voids in big polycrystalline particles (D_{50} ≈ 9.88 μm) increased the electrode's packing density and mechanical strength, as well as preserved the ion routes without spoiling them. During the rolling and cycles, the particle structure also reduced the formation of subcracks induced causing interface damage in the PC-rich electrode. Moreover, according to the enhancement of the dispersal of the conductive additives and buffering of the concentration of stresses, the SC particles are able to further expand the ionic and electronic conductivity of the electrode. Through the combination of PC and SC particles, the bimodal structure still had a capacity retention value of 80.1% through 300 cycles, exceeding the unimodal counterpart with particle cracking and interface resistance suppression compared with common PC electrodes. In short, particle and microstructure joint planning is capable of enhancing all-solid-state lithium battery (ASSLBs) mechanical strength and long-term stability, and it is an effective approach.

There are generally issues occurring within the sulfide-based solid electrolyte (SSE)/high-voltage cathode interface because of mechanical frailty and chemical activity there. Interface layers or surface coatings developed through human effort are effective remedies to these issues and are highly helpful in obtaining the long-term ASSLB behavior. The coatings are passivation layers, able to deter excessive interface reactions, slowing the deterioration process, and maintaining ionic conductivity while preventing electron transmission. Among various methods, oxide-based coatings have shown good results in tests, especially layers made of materials such as $LiNbO_3$ and Al_2O_3. For instance, Liang et al. [3] pointed out that the cathode with a $LiNbO_3$ coating can significantly inhibit the formation of space charge layers and high-resistance interfaces caused by electrochemical potential mismatch. Moreover, hybrid processes, such as a Li_2CO_3/$LiNbO_3$ double-layer positive

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electrode coating, combine cost-effectiveness with enhanced chemical shielding capabilities and have demonstrated potential for commercial applications. However, according to Morchhale et al. [8], coating materials must have low electron conductivity, a wide electrochemical stability window, and strong Li^+ conductivity. Atomic layer deposition (ALD) has been the method of choice for creating ultrathin, conformal coatings on the surfaces of electrolytes in recent years. In a 2023 work by Hood et al. [9], a sulfide-based SSE doubled bulk ionic conductivity and offered chemical passivation against ambient moisture and oxygen on Li_6PS_5Cl (LPSCl) particles coated with ALD-grown Al_2O_3. Despite severe electrochemical and environmental conditions, these coatings maintained the structural and chemical integrity of SSEs. Significant improvements in processability and manufacturability were made possible by the notable ability of coated LPSCl powders to tolerate humid air exposure (>4 hours) with little degradation. Nonetheless, there are still issues with coating consistency, controlling thickness, and preservation during cathode production. These restrictions imply that consistent interfacial performance in sulfide-based ASSLBs depends on the cooperation of coating selection and particle morphology.

Creating a robust, low-resistance interface between the electrode and solid electrolyte in sulfide-based ASSLBs is both a fabrication and a materials problem. Dry electrode fabrication is one of the most scalable processing techniques that has a major influence on the quality of interfacial contact and overall electrochemical performance. For all-solid-state batteries, dry electrode manufacturing provides a scalable solution that circumvents the limitations of slurry-based casting. The use of dry-room conditions (≈ −40 °C dew point), where sulfide electrolytes like Li_6PS_5Cl maintain their structural integrity and ionic conductivity after 24 hours, in contrast to ambient air, is a crucial enabler[10]. This makes processing compatible with lithium-ion production lines and possible outside of gloveboxes. The dry procedure creates dense, homogeneous electrodes by mixing small amounts of binder without the use of solvents, then shearing and hot rolling. Dry processing results in fiber-like binder morphologies and decreased porosity, allowing for free-standing thick electrodes with high areal capacity, in contrast to slurry procedures that increase resistance through binder precipitation. In addition to removing drying processes, roll-to-roll compatible calendaring during the process guarantees accurate thickness control, lowers energy costs, and facilitates pilot-scale production [10]. Cross-sectional scanning electron microscopy (SEM), which shows voids, particle contact, and morphological change, is commonly used to characterize these interface-improving approaches.

3. Analysis of interfacial engineering strategies

Despite significant advancements in electrode density, ionic conductivity, and crack resistance demonstrated by particle and microstructure co-design, there are significant drawbacks that may prevent widespread use. The trade-off between surface reactivity and particle size reduction is a crucial concern. Smaller sulfide electrolyte and cathode active material particles reduce Li^+ transport paths and enhance interfacial area, but they also increase the surface-to-volume ratio. When exposed during manufacture, this accelerates the breakdown of sulfide electrolytes, which are extremely reactive to wet and oxidizing conditions. This can counteract the initial increases in ionic conductivity at the electrode level by accelerating the generation of resistive decomposition products. Furthermore, although exact control over the size distribution is still crucial, smaller SC particles can potentially aid in improving additive dispersion. There are scaling issues with Hong et al.'s bimodal particle design as well. Tight synthesis control is necessary to produce uniform distributions of single-crystalline (SC) and polycrystalline (PC) particles. Although SC particles are an efficient buffer against mechanical stress, scaling up their production may necessitate careful

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process optimization, which could affect cost-effectiveness. PC cracking during cycling may still predominate failure if particle ratios are not optimized, and SC fillers may not be able to sufficiently reduce stress. Long-term stability may be compromised in practice by particle heterogeneity over large electrode sheets, which could lead to localized areas of high impedance and an uneven stress distribution. Some of the inherent issues with sulfide-based ASSLBs are resolved by microstructural engineering, although scalability is called into question due to processing complexity and cost tradeoffs. Whether or not synthesis and mixing techniques can be standardized to yield repeatable particle distributions will determine its long-term effects. Therefore, even though particle co-design shows promise, it might still be more practical for high-end, high-performance cells as opposed to mass-market manufacturing.

Although surface coatings are perhaps the most researched interface engineering technique, they have a number of inherent and useful drawbacks. Controlling thickness is a significant challenge. For oxide coatings like $LiNbO_3$ to permit enough Li^+ transport, they must be incredibly thin—typically only a few nanometers. Overly thick coatings decrease rate capabilities by adding more impedance. Localized cathode surface exposure that is too thin or uneven permits the sulfide electrolyte to decompose, negating any protective properties. Scalable sol-gel or wet-chemical techniques struggle to attain this careful equilibrium. Mechanical durability is another drawback. Cathode materials experience large volumetric changes (up to 8% for layered oxides) during cycling. AlO_3 and $LiNbO_3$ are examples of rigid coatings that can delaminate or break, exposing new, exposed surfaces.Reactive sulfides instantly break down at exposed areas of cracks, creating resistive interphases that propagate throughout the contact. In Ni-rich layered oxides, where structural instability exacerbates the interfacial tension, this issue is more serious. This is somewhat mitigated by hybrid and polymer-based coatings, which provide compliance, but they also present other problems. For example, polymer coatings frequently have poorer ionic conductivity and can constitute bottlenecks for the movement of Li^+. Similarly, stability may be compromised by the dissolution or transformation of carbonate-based coatings (such as Li_2CO_3) over extended cycling. Hood et al. [9] showed that ALD (atomic layer deposition) produces accurate and conformal coatings for AlO_3-coated LPSCl powders from a production standpoint. But because ALD is slow and costly, it is difficult to use on the kilogram-to-ton scale required for cathode powders in commercial batteries. Integration with existing roll-to-roll processes is difficult, even if throughput increases. Additionally, even with premium ALD coatings, transition metals (such Ni and Co) may diffuse into the coating layers at high temperatures during cycling, progressively decreasing the coatings' efficacy. Although coating and artificial interphase design offer effective remedies for chemical instability, there is a basic trade-off between scalability and performance. While industrially feasible techniques (wet-chemistry) introduce unpredictability and diminish efficacy, the most successful coatings (ultrathin, conformal oxides from ALD) are the least scalable. For high-voltage cathodes, this approach is probably going to be crucial, but its continued viability hinges on advancements in scalable, reasonably priced deposition methods that maintain coating consistency and robustness under mechanical stress.

Interfacial contact and manufacturability are directly addressed by process-level techniques like dry electrode manufacturing, but they also have major drawbacks. Although dry electrode manufacturing is very scalable and appealing for energy efficiency, it has drawbacks of its own. Compared to glovebox-level protection, the need for ultra-dry conditions (≈ -40 °C dew point) dramatically raises facility expenses. Before cell construction, LPSCl can decompose into resistive phases due to even slight variations in humidity. Additionally, careful optimization of the shearing and hot rolling is required to prevent microstructural damage or uneven electrode packing. These

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dangers are increased when scaling to wide-format electrodes since even small variations might result in local voids or stress sites that impair cell function. The fact that dry-processed electrodes frequently have low binder contents, which enhance ionic conductivity but degrade mechanical robustness, is another significant drawback. Binder-poor electrodes may lose cohesiveness or delaminate with repeated cycling and external stress, especially at the electrode–current collector interface. Over time, this structural flaw may result in higher impedance, canceling out the initial conductivity improvements. Of the three, process-level techniques are perhaps the most applicable to industry; yet, their drawbacks underscore a conflict between realistic manufacturability and performance enhancement. Whether or not improvements in dry-room infrastructure, binder design, and pressure control can get beyond these obstacles will determine their long-term success.

4. Advancements and future directions

Building on particle and microstructure co-design, artificial coatings, and process-level innovations, this paper explicitly proposes an original combinatorial interfacial engineering strategy. Rather than treating each approach as an isolated solution, I frame the framework as deliberately uniting them into a hierarchical design that addresses chemical, mechanical, and manufacturing challenges simultaneously. At the particle level, bimodal microstructures—large polycrystalline (PC) particles blended with small single-crystalline (SC) ones—achieve both high packing density and reduced microcracking [7]. Complementing this, optimized sulfide electrolyte particle distributions, such as nanoscale Li_6PS_5Cl, minimize voids and enhance ionic percolation across the cathode–electrolyte interface [6]. On top of this optimized architecture, multifunctional surface coatings such as $LiNbO_3$ and Al_2O_3 provide a protective barrier against parasitic side reactions. These ultrathin, conformal layers stabilize the chemical environment at the cathode–electrolyte boundary, suppress the formation of space-charge layers, and reinforce mechanical contact [3,8]. Atomic layer deposition (ALD) demonstrates the highest degree of uniformity and precision, though its expense remains a concern for scale-up. To bridge laboratory techniques with manufacturing, scalable processes such as sol–gel and dry coating methods are increasingly promising alternatives [8,9]. Lastly, solvent-free dry electrode manufacturing is used to consolidate these tailored particles and coatings. Dry manufacturing maintains the inherent stability of sulfide electrolytes, in contrast to slurry-based techniques that introduce moisture and solvent-related deterioration. Additionally, roll-to-roll compatibility and high areal loadings (>6 mAh cm^{-2}) are supported via dry pressing. Binder-free or polymer-assisted approaches further maintain the integrity of ionic and electronic pathways during cycling while ensuring structural uniformity [10]. Taken together, I propose this integrated framework as a multi-scale hierarchy in which particle and microstructure design optimizes ionic transport, surface coatings stabilize interfaces, and dry electrode fabrication ensures scalable manufacturability. Collectively, this original approach is argued to directly address the three critical bottlenecks of sulfide ASSLBs—chemical instability, contact loss, and processing incompatibility [3,5].

The synergy between these strategies arises because each compensates for the limitations of the others to a certain extent. Bimodal particle design reduces microcracking, but the cycling process inevitably exposes fresh reactive surfaces that accelerate degradation. When coupled with conformal surface coatings, these exposed surfaces are immediately protected, preventing electrolyte decomposition while sustaining Li^+ transport [7,8]. Similarly, coatings alone often fracture or delaminate during conventional slurry-based fabrication, yet dry electrode processing eliminates solvent-mediated stresses, thereby preserving coating integrity. Coated sulfide powders also display extended tolerance to ambient exposure, which reduces the cost burden of maintaining ultra-dry

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processing environments [9]. At the microstructural level, small sulfide electrolyte particles enhance ionic conductivity, but their tendency to agglomerate creates non-uniform conduction pathways. Dry pressing counteracts this by dispersing them uniformly within the cathode composite, while larger cathode particles preserve mechanical robustness, producing a dense and continuous conductive network [6,7]. Therefore, I argue that when all three elements—particle engineering, surface modification, and dry processing—are combined, they form a triangular interaction that multiplies rather than merely adds their benefits. Microstructural integrity ensures long-term cycling stability, coatings stabilize the chemical environment, and dry processing guarantees scalable manufacturability. This proposed synergy suggests the possibility of extended cycle life of more than 500 cycles at capacity retention above 80%, while simultaneously enabling practical-level energy densities exceeding 900 Wh L^{-1} [3,6,8].

Although integrated interfacial engineering shows significant promise, several critical challenges remain for future research. One priority is the scalability and cost of advanced coatings. While ALD remains the gold standard for uniformity and performance, its expense makes it impractical for gigafactory deployment. Future work should focus on developing scalable analogues, such as plasma-enhanced chemical deposition, aerosol processes, or sol–gel-based methods, that maintain nanoscale precision without prohibitive costs [8,9]. Another important direction is the real-time characterization of interfacial dynamics. Degradation mechanisms such as space-charge layer formation and crack propagation remain only partially understood. Combining electrochemical impedance spectroscopy with operando cross-sectional SEM could provide unprecedented insight into these evolving interfacial processes and guide the design of next-generation interfaces [3]. Moisture and air stability also remain limiting factors. Sulfide electrolytes degrade rapidly when exposed to trace levels of H_2O or O_2, significantly complicating processing. Coating innovations that extend air stability beyond 24 hours, combined with humidity-tolerant fabrication methods, could drastically reduce dry-room costs and improve industrial feasibility [9]. Additionally, most interfacial strategies focus on cathode optimization, yet Li metal anodes continue to suffer from dendritic growth and poor wetting against sulfide electrolytes. Extending protective coatings and buffer layer approaches to the anode side is essential for realizing symmetric stability across both interfaces [3]. Finally, the transition from laboratory-scale to pilot-scale demonstrations is a pressing need. Current breakthroughs are largely limited to coin-cell testing, which does not reflect the complexity of practical devices. I emphasize that the next step must involve adapting these integrated designs into pouch cells and stacked modules, with careful consideration of stack pressure, thermal management, and mechanical tolerances during operation [10]. Such work would bridge the gap between academic advances and industrial application, ultimately positioning sulfide ASSLBs as viable contenders in electric vehicle and grid-scale energy storage markets.

5. Conclusion

The development of interfacial engineering techniques for sulfide-based all-solid-state lithium batteries (ASSLBs) has been studied in this work, with an emphasis on process-level innovations, surface coatings, and particle/microstructure co-design. The case studies indicated the use of coatings and dry fabrication to stabilize interfaces and improve manufacturability, as well as the potential of tailored microstructures, such as bimodal particle topologies, to decrease microcracking and improve electrode density. These methods show quantifiable improvements in interfacial stability, capacity retention, and ionic conductivity. However, the investigation made clear that each technique had drawbacks that prevent widespread use. In particle co-design, the drawback would be the trade-offs between scalability, surface reactivity, and size reduction. Although surface coatings

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are good at reducing adverse responses, they have problems with thickness control, mechanical durability, and expense, especially when using sophisticated deposition techniques like ALD. Although dry electrode manufacture at process-level provides a scalable route, it requires rigorous environmental control and meticulous optimization to achieve a balance between mechanical cohesion and ionic conductivity. These limitations convey the conflict between industrial feasibility and laboratory creation. A thorough framework that integrates these tactics into a hierarchical architecture was presented in the section on advances and future directions. The strategy of combining integration of solvent-free dry processing, multifunctional surface coatings, and microstructural optimization produces a synergy that improves electrochemical performance definitely more successfully than any one strategy alone. In addition to mechanical and chemical stability, this integration takes into account the manufacturability that is necessary for scaling ASSLBs.

To close the gap between coin-cell research and real-world implementation, future studies must concentrate on scalable coating techniques, real-time interfacial diagnostics, enhanced air/moisture tolerance, and pilot-scale demonstrations. In conclusion, whereas advancements at the particle, surface, and process levels all provide significant contributions, the most obvious route to sulfide-based ASSLBs that are safe, long-lasting, and scalable is provided by their combination into an integrated approach. This strategy has the ability to convert scientific advancements into the practical reality of long-lasting, high-energy batteries for grid storage and electric automobiles. In order to bridge particle design, surface modification, and process-level advances, this research offers an integrated paradigm for interfacial engineering in sulfide-based ASSLBs. It provides a road map for scalable, long-lasting, and high-energy batteries by pointing out both drawbacks and opportunities, directing future developments from lab research to industrial use.

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