

Interfacial Resistance in Solid-State Batteries: Mechanisms and Mitigation

Tianhao Hu

*School of Chemistry and Chemical Engineering, Wuhan University of Science and Technology,
Wuhan, China
haohu6279@gmail.com*

Abstract. Solid-state batteries offer important safety advantages and a feasible route towards higher-energy-density batteries. In practice, however, resistance at electrode-solid-electrolyte interfaces still limits rate capability and cycle life. The main reasons for the interface resistance are insufficient solid-solid contact, chemical reactions at the interface, space-charge layers and grain-boundary structures. Electrolyte chemistry, electrode-electrolyte compatibility, fabrication conditions and cycling-induced mechanical stress are also factors that affect resistance. High interfacial resistance impedes Li^+ transport, increases cell polarisation, and reduces accessible capacity and energy efficiency. The main reduction measures are cathode coatings, artificial anode interlayers, composite solid electrolytes, surface modification and optimisation of the interfacial structure and processing. Further progress will probably require these approaches to be combined. Such combinations must retain low resistance under practical temperatures and stack pressures while meeting electrochemical, cost, and manufacturing requirements. Effective interface engineering should therefore address the dominant failure mechanism, because low initial impedance does not guarantee long-term stability.

Keywords: Solid-state batteries, Interfacial resistance, Solid electrolytes, Lithium-ion transport, Interface engineering

1. Introduction

Traditional lithium-ion batteries have flammable organic electrolytes. Therefore, they are prone to leakage and thermal runaway at high temperatures or after mechanical damage. Their energy density is also approaching the limits of existing materials. Solid-state batteries (SSBs) replace the liquid electrolyte and separator with a solid electrolyte. This structure is compatible with lithium-metal anodes and high-voltage cathodes, and may improve safety and energy density [1]. SSBs have thus become a focus of research for next-generation energy storage.

Commercialisation is still hindered by slow ion transport at the solid-solid interface, thus limiting rate capability and cycle life. Solid electrolytes and electrodes are generally in contact only at discrete points, so the effective contact area is small and interfacial impedance is relatively large. Parasitic reactions can also form resistive interphases and reduce ion transport further. These processes increase cell polarisation and reduce energy efficiency. They may also promote lithium

dendrite growth and increase the risk of local heating and internal short circuiting. Therefore, the origin of interfacial resistance and improved interface-control methods need to be explored [2-4].

Interfacial resistance is a central technical bottleneck in solid-state batteries (SSBs). This review examines the microscopic origins and evolution of resistance, with emphasis on physical contact, electrochemical compatibility, and mechanical stress. It also discusses recent advances in materials modification, interface engineering, and cell-level design. The aim is to support the rational design and practical development of high-performance SSBs.

2. Formation and effects of interfacial resistance

2.1. Formation mechanisms

2.1.1. Insufficient solid-solid contact

A lack of solid-solid contact is a primary source of interfacial resistance in SSBs [2, 4]. As both the electrode and the electrolyte are solids, good contact needs to be realised and maintained by external pressure. Real surfaces are rough; therefore, contact occurs only at a small proportion of the apparent geometric area. The uncovered gaps are not available for Li^+ transport and increase cell polarisation. Repeated volume changes of the electrode during charging and discharging will gradually separate the two phases. This progressive loss of contact further increases interfacial resistance.

The mechanical properties of the solid electrolyte affect the quality and stability of the interface. Oxide electrolytes are relatively rigid and thus more likely to have microscopic voids at the electrode interface. Sulphide electrolytes are softer and can be cold-pressed to increase density; thus, they generally have a larger true contact area. This mechanical advantage, however, is offset by their lower chemical stability. Parasitic reactions can form a resistive interphase and partly negate the benefit gained from improved physical contact [5].

2.1.2. Interfacial side reactions

Chemical incompatibility between the electrode and the solid electrolyte is a second major source of interfacial resistance [6, 7]. Redox reactions during charging and discharging may decompose the electrolyte and form a reaction layer. When the electrochemical stability window of the electrolyte does not cover the operating potential of the electrode, a high-voltage cathode can oxidise sulphide components and promote elemental interdiffusion. Lithium metal can also reduce electrolyte species containing Ge, Ti or Nb. If the decomposition products conduct both electrons and ions, the reaction front can continue to advance into the electrolyte. Sintering may improve the physical contact, but it can also cause lithium volatilisation and form resistive secondary phases. Therefore, an increase in the processing temperature is not always desirable.

2.1.3. Space-charge layer effects

Space-charge layers may be present at the interface of some solid-state materials. When two dissimilar solid materials make contact, the lithium chemical potentials on the two sides are generally unequal. Li^+ therefore redistributes near the interface, forming a nanoscale space-charge layer. The corresponding local electric field can produce a steep gradient in Li^+ concentration and add an energy barrier to cross-interface migration. Their quantitative contribution remains uncertain because space-charge effects are difficult to distinguish from reaction-layer formation, strain, and

defect-mediated transport. Regions with a strong space-charge effect can be buffered by an energy-level-matched layer to reduce the potential gradient at the interface and improve ion transport [8, 9].

2.1.4. Grain-boundary impedance

Grain-boundary impedance is an internal microstructural resistance and not an electrode/electrolyte interface resistance. Both forms of resistance jointly control multiscale Li^+ transport [5], so they are included here. Most inorganic solid electrolytes are polycrystalline. Grain boundaries contain structural discontinuities, local disorder, and a relatively high density of defects. These features may restrict ion migration. If the sintering conditions are not optimised, there will be poor bonding and residual pores at the grain boundaries. The problem is especially prominent in some oxide electrolytes. Optimisation of densification and reduction of grain-boundary defects can therefore lower resistance and improve ionic transport.

2.2. Effects and governing factors

2.2.1. Effects on solid-state battery performance

Interfacial resistance directly limits the transfer of Li^+ at the electrode-solid electrolyte interface. As impedance increases, so does the interfacial overpotential. The cell reaches the charge or discharge cut-off voltage earlier, and some of the theoretical capacity is thus unavailable. Transport limitations are more severe at high charge or discharge rates [2, 3]. Therefore, both fast-charging capability and high-power performance decline.

Electrode volume change during extended cycling may lead to interfacial cracking or delamination. At the same time, poorly ion-conducting interphases formed by side reactions may continue to grow. Both types of degradation reduce the number and continuity of effective Li^+ transport paths. Interfacial impedance therefore increases progressively from its initial value. The resulting polarisation accelerates capacity loss and lowers energy efficiency. This degradation can also increase the risk of local overheating and internal short circuiting [3, 10].

2.2.2. Factors governing interfacial resistance

The structure and properties of the solid electrolyte directly influence interfacial impedance. Oxide electrolytes tolerate high potentials and possess high mechanical strength, but their rigidity makes conformal contact difficult. Sulphide electrolytes are readily densified and often exhibit high ionic conductivity, yet they can decompose at high potentials or against lithium metal. Halide electrolytes are generally more compatible with cathodes, and their instability at low potential often requires isolation from the anode. Polymer electrolytes have good conformability with adjacent surfaces, but their room-temperature conductivity and high-voltage stability are generally relatively poor [5, 11].

Compatibility between the electrode and electrolyte is equally important. High-voltage cathodes can oxidise the unstable electrolyte, and a lithium-metal anode can drive reductive decomposition. Both processes generate interphases with low ionic conductivity and increase transport resistance. Electrode morphology also matters. Particle size, surface morphology, and local roughness influence the effective contact area and the continuity of Li^+ pathways [6, 11].

Fabrication conditions and cycling-induced mechanical stress continuously change the interface. Assembly pressure sets the initial contact area, and sintering temperature controls densification and elemental diffusion. Surface impurities, such as Li_2CO_3 , can also alter wettability. Insufficient

pressure leaves interfacial pores, whereas excessive pressure may fracture the ceramic electrolyte or damage the porous cathode. At an LCO/LLZO interface co-fired at 500 °C, the unmodified specimen had an area-specific impedance of about 8 kΩ cm². This result shows that chemical degradation can dominate even when physical contact appears continuous. Particle expansion and contraction during cycling, lithium plating and stripping, and continuous changes in residual stress shift the contact interface. Higher stack pressure can reduce void formation at the lithium interface, but it may also cause cracking of the ceramic. An increase in temperature can lower some charge-transfer barriers while also accelerating parasitic reactions. Therefore, reported impedance values should only be compared when the temperature, stack pressure, effective area, normalisation method, state of charge and cycling state are the same [3, 10, 12].

3. Strategies for mitigating interfacial resistance

The appropriate mitigation strategy depends on whether the dominant resistance arises at the cathode, at the anode, or within the electrolyte. Cathode-side treatments aim to suppress high-potential side reactions and maintain contact between the active particles and the solid electrolyte. At a lithium-metal anode, good interfacial wetting is needed to avoid void formation, while electron-blocking protection is required to prevent electron entry into the electrolyte. Composite electrolytes also require low-resistance transport pathways between the ceramic and polymer phases. These approaches reduce interfacial resistance by improving physical contact, stabilising interfacial chemistry, and maintaining continuous Li⁺ transport paths.

3.1. Interfacial coatings to suppress cathode-side reactions

Interfacial coatings have been introduced to address the problem of contact-induced side reactions and improve the local ion-transport environment. The basic approach is to add a chemically stable, ion-conducting layer to either the electrode or the solid electrolyte. This protective layer forms a transition region between the two incompatible materials. By limiting direct contact, it suppresses the formation of poorly conducting reaction products and reduces interfacial resistance. If the coating also conducts Li⁺ effectively, it can facilitate ion transfer rather than serving only as a chemical barrier [4, 13].

High-nickel layered oxides and lithium-rich manganese-based cathodes have high oxidation potentials. Direct contact with the sulphide or halide electrolyte will therefore create a large energy mismatch and cause oxidative decomposition. The resulting by-products generally have low ionic conductivity and increase interfacial impedance. Residual surface alkali, lattice-oxygen release and transition-metal migration can also promote sulphide oxidation. Cycling-induced volume changes can crack the protective layer and expose new reactive sites. Typical coating materials are LiNbO₃, Li₃PO₄, Li₂ZrO₃ and LiAlO₂. A good coating should chemically isolate the cathode while remaining permeable to Li⁺. An overly thin layer may not cover the entire active area, whereas an excessively thick layer increases the transport barrier [6, 13].

Müller et al. have studied an LCO/LLZO model cell and found that a Li-Nb-O interlayer reduced the area-specific interfacial impedance from about 8 to 1 kΩ cm². Under the same model system and measurement conditions, Li-Al-O and Li-Ti-O interlayers produced values of 2 and 2.5 kΩ cm² respectively. The Li-Nb-O-modified cell delivered a first-cycle specific discharge capacity of 125 mAh g⁻¹, compared with 26 mAh g⁻¹ for the unmodified specimen. This improvement is consistent with suppressed interfacial reactions in this model system. Capacity nevertheless declined during subsequent cycling, suggesting that the interlayer did not fully prevent mechanically driven cracking

or delamination. These results should not be used to rank thin-film or composite-cathode systems tested with different architectures or under different conditions [12].

Thin-film batteries show a comparable response. After adding a niobate interlayer, the interfacial impedance of LCO/LLZO decreased from about $5 \text{ k}\Omega \text{ cm}^2$ to $53.5 \text{ }\Omega \text{ cm}^2$, a reduction of nearly two orders of magnitude. Given that the contact boundary is relatively clear in a thin-film structure, the result more directly reflects the chemical barrier properties of the interlayer. A typical composite cathode is relatively complicated. The impedance also includes contributions from pores, conductive-additive distribution and particle-particle contacts, so it is generally higher [14].

In a high-nickel NCM811/LGPS system, approximately 1 wt% LiNbO_3 produced a cathode coating about 6.3 nm thick. At 35°C and 0.1 C, the modified material had a specific discharge capacity of 203 mAh g^{-1} . It still provided 136.8 mAh g^{-1} at 5 C and showed good rate performance under the reported conditions. For high-nickel and lithium-rich manganese-based cathodes, coating performance should be evaluated by post-cycling impedance growth, interfacial cracking and stability at a high cut-off voltage; first-cycle capacity alone is not sufficient [15].

3.2. Artificial anode interlayers for improved wetting and electron blocking

Artificial interlayers resemble surface coatings but place greater emphasis on deliberately engineering the structure and reaction path of an interface. They are generally introduced between lithium metal and an inorganic electrolyte to form a stable region for Li^+ transfer. The anode-interface design needs to meet the competing requirements of close contact and electronic insulation. Lithiophilic materials improve lithium wetting and reduce the initial contact resistance, but electronically conducting alloy layers can promote lithium nucleation at electrolyte defects. Electronically insulating layers suppress electron injection but, if their ionic conductivity is too low, will cause additional polarisation. Therefore, many practical designs have combined a lithiophilic component with an ion-conducting, electron-blocking layer. The two components promote wetting and interfacial stabilisation, respectively [2, 3].

Han et al. deposited an approximately 5 nm Al_2O_3 film on a garnet electrolyte by atomic layer deposition. On contact with lithium, the film formed a lithium-containing aluminium oxide phase. This interfacial reaction reduced the room-temperature area-specific impedance from 1710 to around $1 \text{ }\Omega \text{ cm}^2$. The modified symmetric cell then operated stably for 90 h at 0.2 mA cm^{-2} , and there was no significant increase in plating or stripping overpotential. These observations indicate that poor surface wetting accounts for a substantial fraction of the interfacial resistance between garnet and lithium. They also show that nanoscale surface treatment can substantially improve physical contact. Lee et al. designed an Ag/LiF bilayer to improve both the wetting performance and electron-leakage suppression. Ag reduced the lithium contact energy, and LiF served as the electron-blocking component [16, 17].

Compared with a passive coating, an artificial interlayer offers more direct control over interfacial composition, electron transport, and reaction pathways. This design flexibility makes artificial interlayers a promising route for stabilising lithium-metal interfaces.

3.3. Composite solid-electrolyte design

A composite solid-electrolyte structure can also be used to reduce interfacial resistance. A single electrolyte family rarely meets all the mechanical, chemical and transport requirements. Oxides, sulphides and halides therefore have different but complementary limitations. Oxides are relatively stable but generally have poor physical contact. Sulphides have high ionic conductivity and are easy

to densify, but they have poor chemical stability. Combining these phases can integrate their advantages and offset the limitations of a single material [4, 5].

Kuhnert et al. examined Li^+ transport at the interface of LLZTO and poly(ethylene oxide) (PEO). Untreated ceramic-polymer interfaces had relatively large impedances. Plasma activation and subsequent silane grafting formed covalent bonds between the LLZTO surface and PEO. Under the matched-measurement conditions of the study at 20 °C, the modified interface had an area-specific impedance of about 500 $\Omega \text{ cm}^2$ [18]. This value was more than four orders of magnitude lower than the untreated value. The result is specific to the reported LLZTO/PEO architecture and impedance analysis; it should not be considered a general reproducible benchmark. These results indicate that composite-electrolyte performance depends on both ceramic loading and the degree of bonding at the ceramic-surface interface. PEO segmental motion is still limited at low temperatures, and further reductions in interfacial impedance are required.

Gradient and bilayer electrolytes allow the material on each side to be selected for the operating conditions of the adjacent electrode. Oxidation-resistant halides or oxides can be used close to the cathode. Reduction-resistant garnets, appropriately protected sulphides or flexible polymers can be used near the anode. An intermediate layer will connect the Li^+ transport paths and accommodate the differential thermal expansion. This structure does not require a single electrolyte to be compatible with both electrodes. The additional internal interfaces may, however, introduce delamination or space-charge accumulation [3, 5]. Careful compositional and structural design is therefore needed to reduce voids and impedance without adding new interfacial barriers.

3.4. Surface treatment and forming processes

When the high resistance is mainly due to a contamination layer, surface roughness or pressing-induced pores, the electrolyte surface can be treated directly. When exposed to air, garnet electrolytes readily form Li_2CO_3 and LiOH ; both reduce lithium wettability. Mechanical polishing, wet cleaning, heat treatment and acid treatment can remove or modify these surface species. Hot pressing and low-temperature sintering can reduce porosity and expand the area of true contact simultaneously. The above processes are relatively simple; however, their effectiveness and reproducibility require strict control of the treatment parameters [19].

Sharafi et al. used wet polishing and heat treatment to reduce the area-specific lithium/LLZO interfacial impedance from about 400 $\Omega \text{ cm}^2$ to 2 $\Omega \text{ cm}^2$. The interface retained a low impedance for 100 cycles. This treatment directly addressed surface contamination and poor wetting; however, the selected temperature may not be suitable for all garnet compositions. Surface cleaning alone may be inadequate if the electrolyte has significant porosity or electronically conducting grain boundaries. Under these circumstances, lithium can still propagate along internal defects. Thus, the optimisation process should consider electrolyte density, grain size and stack pressure together with surface chemistry [19].

Kim et al. protonated and etched Ta-doped LLZO by acid treatment. At 25 °C, the area-specific lithium/LLZO interfacial impedance was 0.9 $\Omega \text{ cm}^2$ and the tensile strength had increased to 134 ± 7 MPa from 86 ± 14 MPa. The treated electrolyte was then used in an NCM111/Li hybrid full cell with an areal cathode capacity of 3.2 mAh cm^{-2} . At C/2 (about 1.6 mA cm^{-2}), the cell retained 95.0% of its capacity after 1000 cycles and reached a Coulombic efficiency of 99.92% at cycle 1000. A separate all-solid-state cell with a 5 mAh cm^{-2} NCM811/Li₆PS₅Cl composite cathode was operated for more than 1000 cycles at 60 °C and 3 mA cm^{-2} without short-circuiting. Taken together, these cell-specific results demonstrate durable cycling under the reported conditions rather than a universal absence of capacity loss [20].

Assembly pressure and operating temperature will also affect the results of surface treatment. A high-pressure contact generally has a lower initial impedance, but too much pressure may crack a thin electrolyte or alter the pore structure of a composite cathode. Heating can improve lithium wetting and polymer-segment mobility, but it may also accelerate interfacial decomposition. For practical manufacture, surface activation could be integrated with hot pressing or low-temperature sintering. Treatment time, environment, surface roughness and applied pressure also need to be controlled carefully. If a low-resistance interface can only be maintained at a high temperature or large stack pressure, its range of application will be relatively narrow [3, 20].

4. Outlook

Future work should prioritise standardised testing, operando diagnostics, and scalable production. Cathode coatings, artificial anode interlayers and surface modifications have already produced substantial reductions in initial impedance. Many reported values, however, were obtained at elevated temperature, high stack pressure, or in small-area cells. These conditions differ substantially from those expected in practical devices. Therefore, future studies should report and, where possible, standardise the temperature, pressure, effective area and cycling state. Initial impedance should be distinguished from the subsequent rate of impedance growth. Only with the above controls can one determine whether an interface strategy offers real stability or is merely a temporary fix for assembly contact.

Interfacial resistance changes continuously during charging and discharging. Cathode-particle volume change, lithium plating and stripping, and progressive reaction-layer growth all change the initial contact geometry. Measurements taken only before and after cycling are thus often insufficient to determine the main degradation mechanism. Future work should combine operando microscopy, X-ray imaging, and stress measurements to observe the development of cracks and voids, as well as interphase growth, in real time. Coatings, compliant buffer layers or electron-blocking interlayers could then be selected based on the observed failure mode instead of being applied empirically.

Practical production should also consider the uniformity, thickness and cost of the nanoscale interlayer. Vacuum-deposition methods are suitable for laboratory cells, but they are not feasible to apply to large-electrode-area devices. Scalable coating methods and low-temperature sintering are therefore more attractive. The performance should be verified using high-areal-capacity cathodes and thin solid electrolytes, not only in an idealised model cell. An interface strategy will have practical application only if it maintains a low resistance at the common operating temperature and a modest stack pressure.

5. Conclusions

This review has examined the formation, effects, and mitigation of interfacial resistance in SSBs. The principal origins are a small true contact area, interfacial side reactions, space-charge layers and grain-boundary barriers. Their relative contributions depend on the materials system, fabrication conditions and cycling-induced mechanical stress. High resistance increases cell polarisation and reduces rate capability. It reduces the usable capacity and energy efficiency. Different interfacial failure modes require correspondingly different mitigation measures. Cathode coatings suppress high-potential side reactions, and artificial anode interlayers improve lithium wetting and restrict electron leakage. Composite electrolytes and surface modification reduce voids and improve contact. The studies reviewed here show that a suitable interlayer or surface treatment can reduce

interfacial impedance by one or more orders of magnitude. Low initial impedance does not necessarily persist during repeated cycling. Strategy selection should therefore begin by identifying the dominant failure mechanism and then choosing the most appropriate coating, buffer layer, composite structure, or processing method.

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