



Beyond ionic conductivity: rethinking the chemo-mechanical design for next-generation solid-state energy storage

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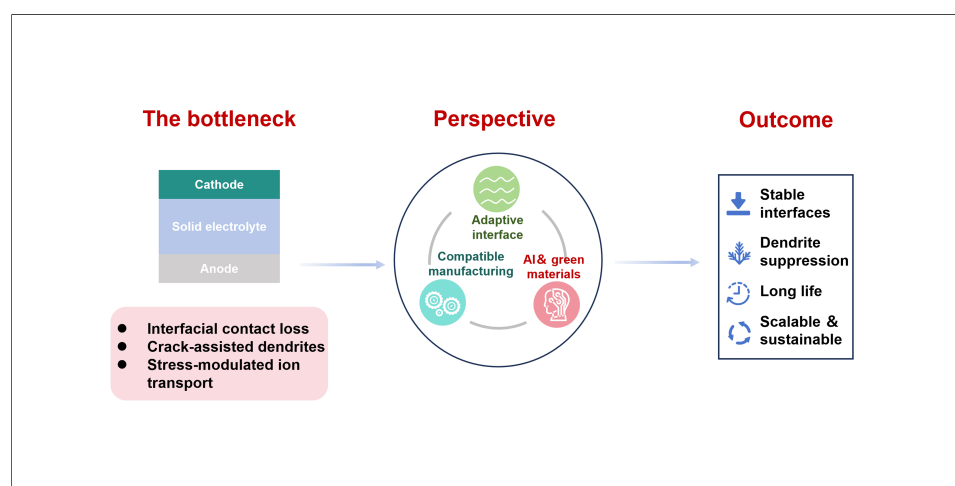
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INTRODUCTION

Over the past decade, in solid-state battery research, the academic community has developed an almost “metric-driven obsession” with the ionic conductivity of solid electrolytes. From sulfide and oxide electrolytes to emerging halide-based systems, the ionic conductivity of solid electrolytes has increased from approximately 10^{-4} S·cm⁻¹ to 10^{-2} S·cm⁻¹, and in some cases has even surpassed that of conventional liquid electrolytes^[1,2]. However, a puzzling phenomenon remains: even when extremely high bulk ionic conductivity is achieved, all-solid-state batteries still suffer from severe capacity fading, rapid increases in internal resistance, and even short-circuit failure during practical cycling^[3-7].

This indicates that ionic conductivity is no longer the sole, or even the dominant, bottleneck to the practical application of solid-state batteries. A solid-state battery is not simply an assembly of materials, but rather a complex chemo-mechanical coupling system^[8]. From this perspective, future breakthroughs are unlikely to come



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merely from discovering materials with higher ionic conductivity. Instead, greater emphasis should be placed on re-examining and rationally designing the coupled “mechanical-chemical” mechanisms within the battery.

CORE ORIGIN OF FAILURE: OVERLOOKED CHEMO-MECHANICAL INSTABILITY

Unlike liquid-based batteries, in which electrodes can be effectively wetted by fluid electrolytes and volume variation can be buffered by their intrinsic fluidity, such physical “soft buffering” is essentially absent in solid-state systems^[9–12]. The failure of solid-state batteries can therefore be understood from three interrelated dimensions [Figure 1]^[13,14]. As illustrated in Figure 1A, the large volume variation of Si can progressively disrupt the intimate contact with $\text{Li}_6\text{PS}_5\text{Cl}$ and promote heterogeneous interfacial reactions, resulting in impedance growth and capacity degradation. Figure 1B further indicates that lithium penetration through solid electrolytes is not solely a mechanical process but can be accompanied by local electrochemical corrosion and crack evolution. Similar chemo-mechanical effects have also been observed in Ni-rich cathodes with different particle microstructures^[15]. Polycrystalline particles are more susceptible to intergranular cracking during calendaring and cycling, whereas single-crystal particles better preserve their structural integrity, although anisotropic lattice strain and interfacial contact degradation may still occur. Collectively, these examples demonstrate that chemical decomposition, mechanical damage, and nonuniform ion transport are strongly coupled during the failure of solid-state batteries. First, during ion insertion and extraction, active materials, such as high-nickel layered cathodes or silicon anodes, undergo severe lattice strain^[16–19]. The resulting periodic volume expansion and contraction can induce microcrack formation at the solid-solid interface, leading to interfacial contact loss, a sharp increase in impedance, and a nonuniform distribution of local current density. Second, although high-modulus ceramic electrolytes have traditionally been considered effective in physically blocking lithium dendrite propagation, this view involves an inherent mechanical paradox^[20,21]. A high modulus is often accompanied by high brittleness, and minor processing defects or cracks generated under cyclic stress may instead become low-resistance pathways for dendrite penetration. Third,

local mechanical stress can alter the distribution of chemical potential at the interface, thereby fundamentally affecting interfacial ion-transfer kinetics. Such strong mechano-electrochemical coupling is difficult to accurately capture using conventional static electrochemical models.

FUTURE PERSPECTIVES ON DESIGN STRATEGIES

To address the above challenges, the design logic of solid-state batteries should be fundamentally reconsidered from the following three dimensions [Figure 2].

Construction of dynamic adaptable interface

An ideal interface should not be limited to a single “rigid” or “soft” characteristic; instead, viscoelasticity and a gradient in modulus should be incorporated^[22–24]. Accordingly, interface design may be considered from the following perspectives. Through *in situ* polymerization and hybrid design, the fluidity of polymer electrolytes and the high mechanical strength of inorganic electrolytes can be combined to construct an *in situ* composite interface. Such an interface is expected to self-adjust during cycling, similar to an adhesive layer, thereby maintaining the integrity of electrochemical contact^[25–27]. In addition, a gradient-modulus interlayer can be designed between the electrode and the electrolyte, allowing the modulus to transition gradually across the interface. In this way, local stress concentrations generated during cycling can be absorbed and dissipated.

However, dynamic adaptability must be balanced with ionic transport and chemical stability. Excessively strong interactions can restrict polymer-chain motion and increase interfacial resistance, whereas overly weak interactions may be insufficient to maintain contact during repeated deformation. Moreover, polar groups such as hydroxyl, carbonyl, amino, and catechol can enhance adhesion and ion coordination but may also react with lithium metal, promote the decomposition of sulfide electrolytes, or undergo oxidation at high-voltage cathodes^[23]. Therefore, asymmetric or composition-gradient interlayers may be more practical than a universal coating. Their implementation also requires careful control of interlayer thickness, precursor reactivity, curing conditions, coating uniformity, and large-

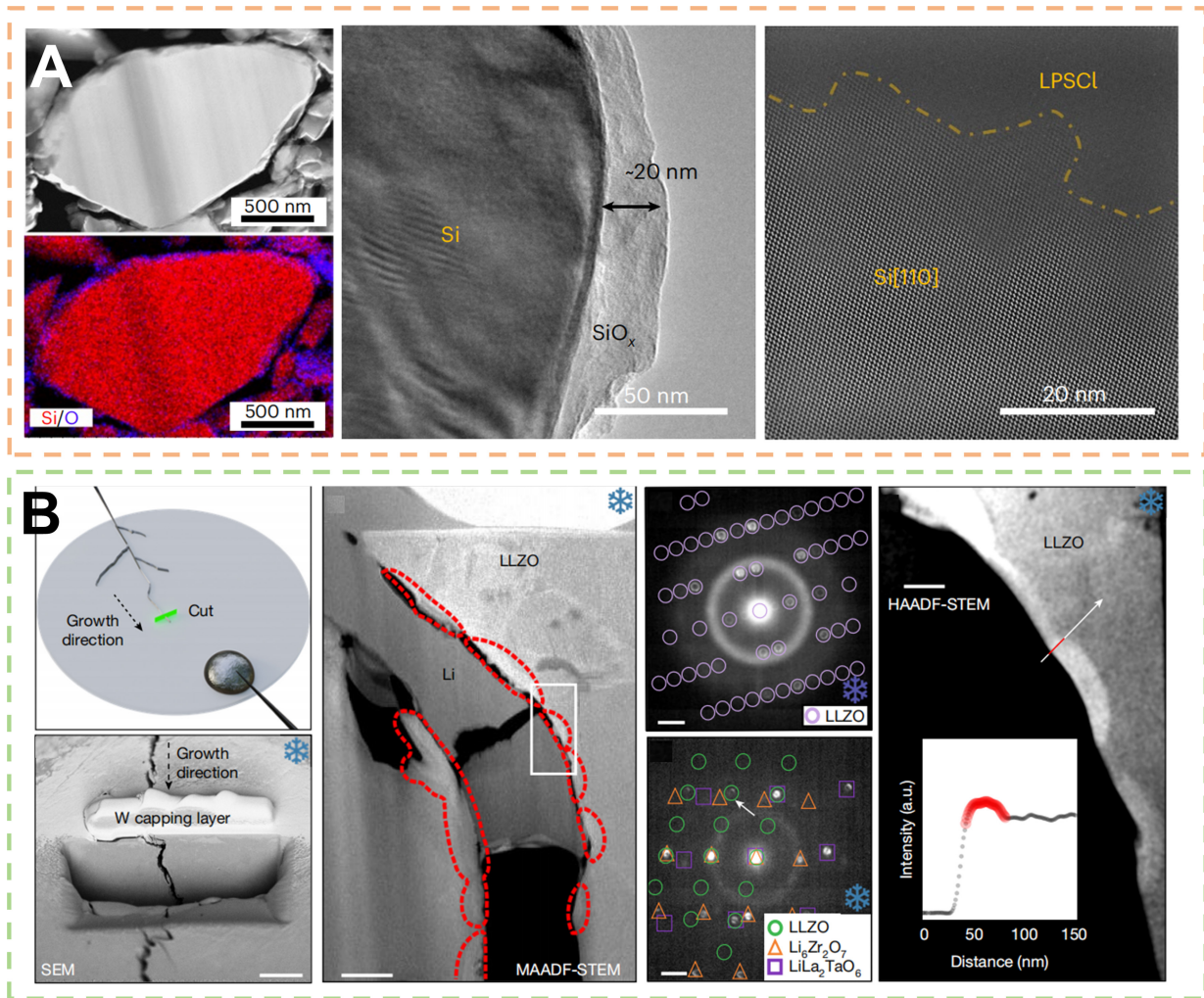


Figure 1. (A) Chemical stability of composite Si/Li₆PS₅Cl anodes^[13]. Adapted with permission. Copyright 2024, Springer Nature; (B) Dendrite-associated degradation visualized by cryogenic electron microscopy^[14]. Adapted with permission. Copyright 2026, Springer Nature. LPSCl: Li₆PS₅Cl lithium argyrodite solid electrolyte; LLZO: Li₇La₃Zr₃O₁₂ lithium lanthanum zirconium oxide; MAADF-STEM: medium-angle annular dark-field scanning transmission electron microscopy; HAADF-STEM: high-angle annular dark-field scanning transmission electron microscopy; SEM: scanning electron microscopy.

area manufacturability. Future adaptive interfaces should therefore be evaluated using coupled metrics, including adhesion strength, stress-relaxation capability, ionic conductivity, electrochemical stability, interfacial impedance, and processing scalability.

Mechanical compatibility in manufacturing processes

The high-pressure cold-pressing procedures commonly used for pellet fabrication in laboratories cannot be readily translated into scalable manufacturing. Therefore, future engineering efforts should place greater emphasis on mechanical compatibility during processing. Solid electrolytes must be fabricated into ultrathin and flexible membranes, typically with a thickness below 30 μm , so that high energy density can be maintained^[28–31]. At such a small thick-

ness scale, however, retaining sufficient mechanical strength and suppressing pinhole defects during manufacturing become central issues in process design. In addition, dry-electrode technology should be further developed to eliminate the need for solvent recovery. Meanwhile, a three-dimensional mechanically supportive electrode network can be constructed through fibrillated binders, thereby improving structural integrity during electrode fabrication and battery cycling^[32–34]. Nevertheless, binder distribution, electrode density, ion-transport continuity, and compatibility with solid electrolytes must be carefully balanced. Scalable manufacturing therefore requires the simultaneous optimization of membrane thickness, defect density, interfacial contact, stack pressure, and production uniformity rather than the isolated improvement of a single

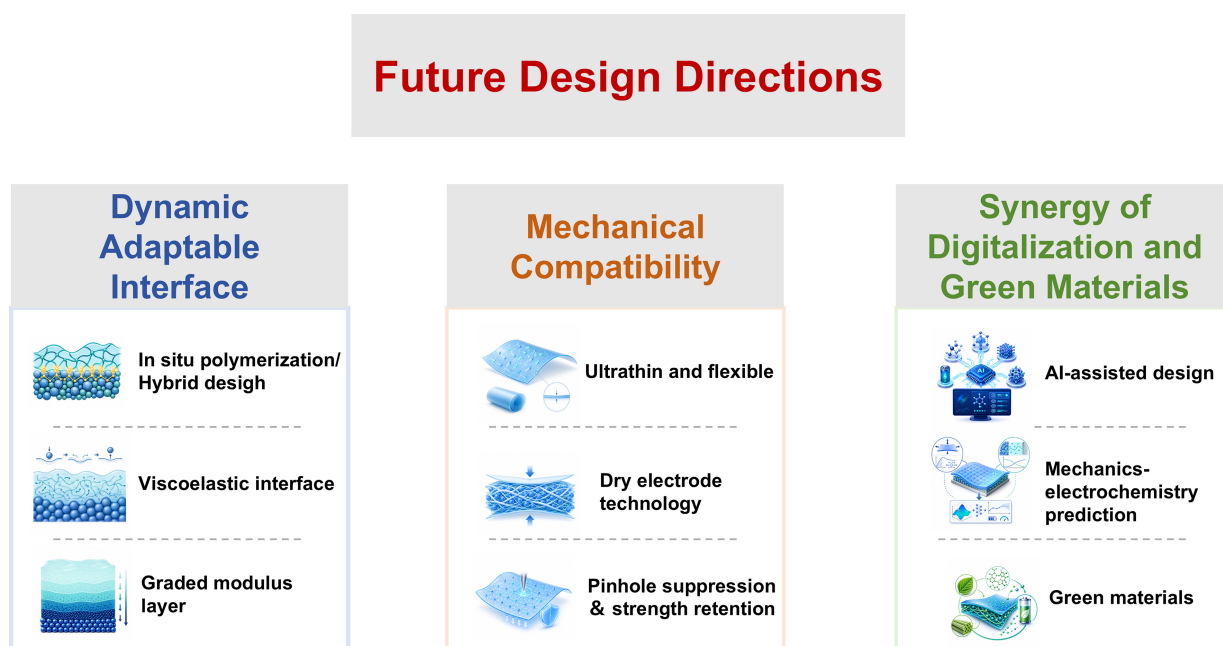


Figure 2. Future design directions for solid-state batteries from a chemo-mechanical perspective.

mechanical parameter.

Synergy of digitalization and green materials

In addition, the synergy between digitalization and green materials is expected to provide a more effective route toward high-performance solid-state batteries. AI-assisted design can be employed to predict the nonlinear relationships between the mechanical properties of materials, such as Young's modulus and fracture toughness, and their electrochemical stability, thereby accelerating the screening of new materials with superior mechanical performance^[35,36].

From a sustainability perspective, biomass-derived polymers can serve as mechanically compliant and chemically functional interfacial regulators^[37,38]. Cellulose derivatives provide robust film-forming frameworks, lignin offers aromatic backbones and phenolic groups, tannin-based materials possess abundant catechol or pyrogallol groups for adhesion and ion coordination, and chitosan can form dynamic hydrogen-bonding networks through its amino and hydroxyl groups. Nevertheless, these materials also face challenges, including moisture sensitivity, compositional variability, limited intrinsic ionic conductivity, and insufficient electrochemical stability. Chemical derivatization, cross-linking, hydrophobic modification, and hybridization with inorganic

fillers may therefore be required. Candidate biopolymers should be evaluated according to coupled criteria, including adhesion strength, mechanical adaptability, ionic conductivity, electrochemical stability, moisture resistance, and compatibility with scalable processing, rather than being selected solely on the basis of their renewable origin.

CONCLUSIONS AND FUTURE PERSPECTIVES

The success of solid-state batteries should not be regarded merely as a triumph of electrochemical theory, but rather as the result of deep integration between mechanics and materials science. Future research should shift its focus from the sole pursuit of "ion migration rate" toward the mechanical durability of the interface. In particular, multiphysics models coupling electrical, chemical, mechanical, and thermal fields should be integrated with advanced *in situ* and operando characterization. For example, operando X-ray tomography can reveal crack propagation and interfacial contact loss, *in situ* X-ray diffraction can track lattice strain and phase evolution, and integrated pressure or strain sensors can monitor stress changes during cycling. These experimentally measured parameters can be used to calibrate and validate models linking local stress, ion transport, and interfacial degradation, thereby enabling failure prediction and rational interface design. Such an integrated approach may help

bridge the gap between laboratory-scale demonstrations and practical solid-state batteries.

DECLARATIONS

Authors' contributions

Proposed the concept and wrote the manuscript: Xu, L

Literature analysis, figure preparation, manuscript discussion, and revision: Xu, L.; Tu, H.; Wang, H.; Yang, X.; Li, C.

All authors approved the final version of the manuscript.

Availability of data and materials

Not applicable.

AI and AI-assisted tools statement

During the preparation of this manuscript, the AI tool ChatGPT (GPT-5.3 Instant, released 2026-03-03) was used for language polishing and to assist in generating several schematic elements in [Figure 2](#) for conceptual visualization. These schematic elements do not represent experimental data or results. The tool did not influence the study design, data collection, analysis, interpretation, or the scientific content of the work. All AI-assisted textual and graphical content was carefully reviewed, edited, and verified by the authors. All authors take full responsibility for the accuracy, integrity, and final content of the manuscript.

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Conflicts of interest

Xu, L. serves as an Editorial Board Member of the journal *Advanced Energy Conversion* but is not involved in any stage of editorial processing, notably reviewer selection, manuscript handling, or decision-making. The other authors declare that they have no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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