

Interfacial instabilities in halide-based solid-state batteries

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All-solid-state batteries have emerged as a promising technology for energy storage, offering improved safety and potential for higher energy density. Halide-based batteries have gained popularity due to the advantageous characteristics of electrolytes, including decent ion conductivity, good formability, high-voltage stability, and moisture resistivity. Despite the impressive cycle life observed in halide-based batteries under high stack pressures or at elevated temperatures, poor cathode–electrolyte stabilities still pose a significant challenge that results in rapid capacity decay under ambient temperature and low pressure. The poor stability at the halide–anode interface further limits the choice of electrode materials for high-energy applications. This article presents a review of interfacial instability in halide-based solid-state batteries, addressing both the chemical, electrochemical, and mechanical origins of these instabilities at the cathode–electrolyte and anode–electrolyte interfaces. We also discuss state-of-the-art approaches to mitigate interfacial instabilities and highlight their limitations. Finally, we propose perspectives and future directions for resolving interfacial instabilities in halide-based solid-state batteries.

Introduction

Lithium-ion batteries have revolutionized various industries, from small household appliances to large-scale energy storage since their commercialization in 1991. As the demand for electric vehicles (EVs) rapidly grows, concerns regarding the energy density and safety of conventional liquid electrolyte batteries have arisen. Solid-state batteries have emerged as a potential solution, offering improved safety and higher energy density. The choice of solid electrolyte is critical in solid-state battery development, with four major categories being widely explored: polymers, oxides, sulfides, and halides.¹ Polymer electrolytes exhibit good formability but typically have low ionic conductivity at room temperature.² Oxide electrolytes provide excellent atmospheric air stability but suffer from poor solid-solid contact, mechanical brittleness, and energy-intensive processing.³ Sulfide electrolytes offer good formability and high ionic conductivities but face challenges in compatibility with high-voltage cathodes and are susceptible to hydrolysis in a humid environment.⁴ In contrast, halide electrolytes exhibit decent Li⁺ conductivity, high-voltage stability, good deformability, unique moisture stability, and potentially enabling scale-up manufacturing as shown in Figure 1a.⁵

Halide electrolytes started to become popular in the solidstate battery community in 2018 when Asano et al. first synthesized Li₃YCl₆ and Li₃YBr₆ electrolytes using mechanochemical methods.⁶ These materials exhibited high roomtemperature ionic conductivity (>1 mS cm⁻¹) and excellent electrochemical performance with 4 V active material LiCoO₂ (LCO). These exceptional properties intrigued researchers to further investigate on halides. Mo et al. presented thermodynamic electrochemical windows of Li-M-X ternary compounds in various halide, oxide, and sulfide systems in Figure 1b.⁷ Although fluoride electrolytes offer a wide electrochemical window, their room-temperature ionic conductivities are typically low due to the formation of $LiMF_4$, which impedes Li-ion transport.⁸ On the other hand, iodine and bromide counterparts are prone to oxidative decomposition and do not exhibit superior stabilities compared to sulfides. In contrast, chlorides exhibit desirable anion chemistries, offering both high ionic conductivity and excellent voltage stability, making them a central focus within the halide electrolyte family. Additionally, chlorides can be synthesized using facile methods, such as the wet chemistry approach reported by

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Sun et al., which enables cost-effective mass production while ensuring precise control over product quality.⁹ Common halide electrolytes and their preparation methods are summarized in **Table 1**.

Despite the progress made, practical implementation of these materials still faces challenges in real-world applications. Most studies on halide solid-state batteries require operation at high stack pressures and low current densities.¹⁰⁻¹⁴ Other approaches to achieving stable cycling performance involve reducing the cathode active material fraction or lowering the charge cutoff voltage.^{10,15} However, the current research on halide electrolytes tends to lack a fundamental and systematic understanding of the underlying mechanisms. Therefore, a comprehensive review is necessary to provide a thorough understanding of these instabilities and address the existing knowledge gaps in the field.

In this article, we delve into the instabilities at the cathode-electrolyte and anode-electrolyte interfaces, exploring both their chemical and mechanical origins. We provide an overview of the strategies employed to address these interfacial instabilities, highlighting their respective limitations. Finally, we propose prospective insights and future directions aimed at effectively mitigating interfacial instabilities in halide-based solid-state batteries.

Cathode-halide interface instability

Most chloride and fluoride solid electrolytes offer the advantage of high-voltage stability, enabling direct cathode–electrolyte contact without triggering side reactions. Initially, the instability of cathodes in halide cells was not a significant concern in early studies due to the thermodynamic stability exhibited by most chlorides up to 4.4 V versus Li⁺/Li, and even higher for fluorides (beyond 6 V) (Figure 1b).^{16,17} However, recent studies have revealed specific challenges associated with cathodes, including electrochemical side reactions with other cell components and mechanical instability resulting from limited solid–solid contact and volume changes, leading to void formation (**Figure 2**a).

Chemical instability at the cathode–halide interface was highlighted in a study by Kang et al.¹⁸ They observed the chemical decomposition of Li_3InCl_6 at $LiNi_{0.8}Ni_{0.1}Co_{0.1}O_2$



Table 1. Overview of common name electrolytes properties.								
Formula	Synthesis Method	σ (mS cm ⁻¹)	References					
Li ₃ InCl ₆	Ball milling + annealing (260°C)	1.49	16					
Li ₃ InCl ₆	Freeze-drying	-	49					
Li ₃ YCl ₆	Ball milling + annealing (550°C)	0.51	6					
Li ₃ YCl ₆	Wet chemistry	0.35	9					
Li ₃ YCl ₆	Ball milling	0.40	10					
Li ₃ YBr ₆	Ball milling + annealing (550°C)	0.72	6					
Li ₃ YBr ₆	Wet chemistry	1.09	9					
Li ₃ YBr _{5.7} F _{0.3}	Solid-state reaction (950°C)	1.80	42					
Li _{2.25} Zr _{0.75} Fe _{0.25} Cl ₆	Ball milling + annealing (500°C)	0.98	11					
$Li_2In_{1/3}Sc_{1/3}CI_4$	Solid-state reaction (650°C)	2.00	14					
Li ₃ ScCl ₆	Wet chemistry	1.25	9					
Li ₂ Sc _{2/3} Cl ₄	Solid-state reaction (650°C)	1.50	17					
Li ₃ ErCl ₆	Wet chemistry	0.41	9					
$Li_{2.60}Yb_{0.60}Hf_{0.40}Cl_{6}$	Ball milling + annealing (400°C)	1.50	12					
Li _{2.556} Yb _{0.492} Zr _{0.492} Cl ₆	Solid-state reaction (450°C)	1.58	13					
Li _{0.388} Ta _{0.238} La _{0.475} Cl ₃	Ball milling	3.02	45					

(NMC811) cathode surface during aging at elevated temperatures. The degradation of composite cathodes was more pronounced at low state of charge (SOC), while high SOC aging had minimal impact on capacity retention. Time-offlight secondary ion mass spectrometry (ToF-SIMS) depth profiles showed an increase in fragments associated with Li₃InCl₆ decomposition (InO₂⁻, LiInO₂⁻) with aging (Figure 2b), while those from unreacted species (InCl₂⁻, NiO⁻) decreased.¹⁸

Decomposition of Li₃InCl₆ could also occur in the presence of other cell components, as reported by Tarascon et al. (Figure 2c). They observed an increase in anodic current during the oxidation of Li₃InCl₆ and vapor-grown carbon fiber (VGCF) mixed with Li₆PS₅Cl electrolyte, leading to the formation of In_xCl_y as the major decomposition product.¹⁹ Interestingly, subjecting the system to high potential initially resulted in a rapid increase in interfacial resistance within the first 10 h, followed by a slower progression, suggesting a self-limiting decomposition process referred to as "aging."¹⁴ This aging process facilitates the formation of a stable cathode-electrolyte interphase with low electronic conductivity (Figure 2d). Additionally, the aging process helps restore intimate particle contact after the volume change of the active material in the initial cycle,¹⁵ eliminating the need for a surface coating layer that hinders electron transport (Figure 2e). Mechanical instability poses a significant challenge to cathode performance, necessitating high stacking pressure during operation (Table II).¹⁰⁻¹⁴ Bruce et al. demonstrated that contact loss between the cathode active material (CAM) and solid electrolyte plays a significant role in the capacity fade of halide-based batteries.¹⁵ The primary cause is the volume change of CAM, which creates surrounding voids that impede Li-ion transport. To address this, a lower cutoff voltage of 4.2 V (Figure 2f) was implemented, considering that LiNi_{0.83}Mn_{0.06}Co_{0.11}O₂ exhibit a smaller volume change of 2.5% at 4.2 V compared to 6% at 4.4 V. Lowering the cutoff voltage led to a significant reduction in voids in the composite cathode.

Mitigating void formation can also be achieved by decreasing the CAM fraction in composite cathodes. Jung et al. reported improved CAM-electrolyte contact and cycling performance when the CAM fraction decreased from 70.9% to 59.3% (Figure 2g).¹⁰ Reduction in the CAM fraction decreases overall volume change and improves interfacial contact. Moreover, cross-sectional scanning electron microscope (SEM) images indicated the presence of internal cracks in the active material resulting from repeated structural transformation, particularly associated with the detrimental H2-H3 phase transition at around 4.1 V (versus Li/Li⁺), which also contributes to capacity decay in solid-state batteries.

It's worth noting that although the majority of research concerning halide batteries employs cathodes made from transition-metal oxides to fully leverage their high-voltage stability advantage, recent studies have indicated the potential compatibility of halides with alternative cell chemistries. Notably, Sun et al. showcased the operation of Li-O₂ batteries at 4.5 V using Li₃InCl₆-infused carbon nanotubes, capitalizing on halide's exceptional anodic stability.²⁰ For other conversion-type electrodes such as S or Se, which operate at lower cutoff voltages, the incorporation of specific central metal elements such as holmium becomes necessary.^{21,22} Furthermore, due to the substantial volume changes associated with the conversion reaction, maintaining interfacial contact demands elevated stack pressures of up to 300 MPa, thereby introducing additional complexities to solid-state batteries.²²

In summary, cathode instability in the halide electrolyte encompasses both electrochemical and mechanical aspects.

INTERFACIAL INSTABILITIES IN HALIDE-BASED SOLID-STATE BATTERIES



Figure 2. Halide electrolyte–cathode instabilities. (a) Schematic illustration of electrochemical and mechanical instability at the cathode– electrolyte interface. CEI, cathode–electrolyte interphase. (b) Time-of-flight–secondary ion mass spectrometry depth profiling results for the composite cathodes exposed under different aging conditions. Reprinted with permission from Reference 18. © 2023 Wiley. (c) Cyclic voltammetry measurement at 0.01 mV s⁻¹ upon 10 cycles between 2 and 3.9 V versus Liln/In. Inset shows the assembled cell configuration. VGCF, vapor-grown carbon fiber. Reprinted with permission from Reference 19. © 2022 American Chemical Society. (d) Leakage current during the aging of LiNi_{0.85}Co_{0.1}Mn_{0.05}O₂-halide at a constant voltage of 4.8 V versus Li⁺/Li after the first charge. Reprinted with permission from Reference 14. © 2022 Springer Nature. (e) Schematic illustration of ionic and electronic conduction percolation within a cathode composite of LiNbO₃-coated LiNi_{0.85}Co_{0.1}Mn_{0.05}O₂ cathode with a sulfide solid electrolyte (SE), and bare cathode with a chloride SE. Reprinted with permission from Reference 14. © 2022 Springer Nature. (f) Reconstructed 3D structures of the cathode composite at cutoff 4.4 V (left) and cutoff 4.2 V (right). Reprinted with permission from Reference 15. © 2022 Elsevier. (g) Cross-sectional scanning electron microscope images of LiNi_{0.88}Co_{0.11}Al_{0.01}O₂/Li₃YCl₆ electrodes with (j) pristine, (ii) 29.1 wt% Li₃YCl₆, and (iii) 40.7 wt% Li₃YCl₆. Reprinted with permission from Reference 10. © 2021 Wiley.

Electrochemical instability not only occurs when the cell potential exceeds the electrochemical stability window (ESW) of halides, but also originates from the decomposition of other cell components to form halide-reactive compounds. The decomposition can be controlled through an aging process that kinetically stabilizes the interface, preventing the formation of halide-reactive compounds. Meanwhile, mechanical instability arises from the volume changes in the active materials. External mitigation strategies involve applying high stack pressure, whereas internal approaches focus on reducing cathode volume change through lower cutoff voltages or decreased CAM fractions. However, it is important to note that these approaches often involve a tradeoff between cycling stability and cell-level energy density. Achieving a balance between energy density and cycling stability is a critical consideration in the development of halide-based batteries.

Anode-halide interface instability

The instability of the anode–halide interface primarily arises from the use of a Li metal anode. Although the Li metal anode offers high energy density with its low electrochemical potential (-3.04 V versus standard hydrogen potential) and high theoretical specific capacity (3860 mA h g⁻¹),²³ it also leads to a highly reductive interface and significant volume changes during cycling.^{24,25} **Figure 3**a illustrates the (electro)chemical and mechanical instability at the anode–halide interface. Halide decomposition occurs chemically on the Li metal surface, even in the presence of a protective layer. Additionally, the substantial volumetric change of lithium results in inhomogeneous stripping/plating and eventually leads to electrolyte penetration.

Most halide electrolytes exhibit cathodic decomposition at an electrochemical potential well above 0 V, which contrasts their good stability against oxidation. First-principles

References	Active Material*	Solid Electrolyte	Pressure (MPa)	Temp (°C)	Retention		
10	p-LiNi _{0.88} Co _{0.11} Al _{0.01} O ₂	Li ₃ YCl ₆	70	30	77.4% (200 cycles)		
	s-LiNi _{0.88} Co _{0.11} Al _{0.01} O ₂	Li ₃ YCl ₆	70	30	96.8% (200 cycles)		
11	s-LiNi _{0.88} Co _{0.11} Al _{0.01} O ₂	Li ₂ ZrCl ₆	70	30	91.3% (100 cycles)		
	LiCoO ₂	Li ₂ ZrCl ₆	70	30	90.5% (100 cycles)		
12	s-LiNi _{0.88} Co _{0.11} Al _{0.01} O ₂	Li _{2.6} Yb _{0.6} Hf _{0.4} Cl ₆	70	30	83.6% (1000 cycles)		
13	LiCoO ₂	$Li_{2.556}Yb_{0.492}Zr_{0.492}Cl_{6}$	100	25	82.1% (50 cycles)		
	LiNi _{0.83} Co _{0.12} Mn _{0.05} O ₂	Li _{2.556} Yb _{0.492} Zr _{0.492} Cl ₆	100	25	78.3% (50 cycles)		
14	LiCo0 ₂	Li ₂ In _{1/3} Sc _{1/3} Cl ₄	250	50	100% (500 cycles)		
	LiNi _{0.85} Co _{0.1} Mn _{0.05} O ₂	Li ₂ In _{1/3} Sc _{1/3} Cl ₄	250	25	80% (3000 cycles)		
	LiNi _{0.85} Co _{0.1} Mn _{0.05} O ₂	Li ₂ In _{1/3} Sc _{1/3} Cl ₄	100	25	89.1% (695 cycles)		
	LiNi _{0.6} Co _{0.2} Mn _{0.2} O ₂	Li ₂ In _{1/3} Sc _{1/3} Cl ₄	250	25	92.8% (320 cycles)		
22	Se	Li ₃ HoCl ₆	300	25	65.4 (750 cycles)		

Table II. Stack pressure, temperature, and cycle life used in reported halide all-solid-state batteries.¹²⁻¹⁶

*p: polycrystalline; s: single crystalline.

calculations by Mo et al. reveal the electrochemical stability windows (ESWs) of Li₃YCl₆ and Li₃InCl₆ to be 0.62–4.21 V and 2.38–4.26 V versus Li⁺/Li, respectively.⁷ The phase equilibria in Figure 3b shows that a typical halide electrolyte Li₃MX₆ (M=metal, X=halide) decomposes to LiX, metallic M, and M₂X₃ at low potential. Zeier and Janek et al. have experimentally confirmed the rapid decomposition of Li₃InCl₆ and Li₃YCl₆ to LiCl and metallic In or Y through *in situ* x-ray photoelectron spectroscopy (XPS).²⁶ Furthermore, such decomposition is spontaneous and continuous, as evidenced by a progressive increase in interfacial impedance over time. It occurs due to the formation of an electrically conductive decomposition product, known as a mixed ion/electronconducting interphase,²⁷ which allows for continuous reaction by consuming Li₃MX₆ and Li metal until their depletion.

To address the cathodic instability of halide electrolytes, lithium alloys such as Li-In can be used instead of pure Li metal. Lithium alloys provide an attractive alternative for establishing a stable electrolyte-electrode interface that enables long-term cycling for solid-state batteries.^{28,29} Typically, alloying Li with other metals increases electrochemical potential and mitigates the decomposition of halides. Additionally, certain Li alloys exhibit higher lithium diffusivity compared to pure Li, facilitating uniform stripping and plating within the electrode.^{30–32} Among various lithium alloys, the Li-In alloy stands out as a popular choice for halide-based batteries due to its mechanical ductility and a constant redox potential (0.62 V versus Li⁺/Li) over a wide stoichiometric range.^{29,33} Asano et al. demonstrated a stable cycling performance of 100 cycles at 0.135 mA cm⁻² with a Li₃YCl₆ and Li-In anode.⁶ However, it should be noted that using a Li-In alloy as the anode significantly reduces the operating potential of the full cell by 0.62 V, leading to a compromised energy density.

Another strategy to resolve anode instability is to avoid direct Li-halide contact. Introducing a protection layer at the Li-halide interface facilitates interfacial Li-ion transport and mitigates undesirable side reactions without sacrificing cell voltage. These protective layers should possess

characteristics such as ionic conductivity, electronic insulation, electrochemical stability, and strong wetting with both halides and Li metal.²³ The argyrodite electrolyte Li₆PS₅Cl is widely investigated for this purpose. Despite having a narrower thermodynamic ESW, Li₆PS₅Cl remains kinetically stable against Li metal through its self-passivating decomposition products, including Li₂S, Li₃P, and LiCl.^{34,35} Qu et al. illustrated the role of a Li₆PS₅Cl layer in Figure 3c, where it selectively allows Li-ion transport while blocking electron transport.³⁶ The mixed ion and electron-conducting nature of the Li/Li₃YCl₆ interphase results in a very thick decomposition layer, while the electron-insulating nature of the Li/ Li₄PS₅Cl interface generates a self-limiting solid-electrolyte interphase (SEI) layer with a moderate thickness. As a result, the Li|Li₆PS₅Cl|Li₃YCl₆|Li₆PS₅Cl|Li symmetric cell showed low polarization overpotential at 100 mV over 1000-h cycling.

The approach of using a protective layer still faces several challenges. First, not all halide electrolytes are compatible with the Li₆PS₅Cl layer. Recent findings by Zeier et al. revealed chemical incompatibility between Li₆PS₅Cl and Li₃InCl₆, as confirmed by ToF–SIMS and scanning electron microscopy assisted by focused ion beam (FIB-SEM). This analysis showed the presence of an indium sulfide-rich region at the contact area between the halide and sulfide materials (Figure 3d).³⁷ To address this interfacial challenge, Tarascon et al. proposed a new surface engineering strategy. They suggested depositing a nanometer-thick Li₃PO₄ coating at the surface of a Li₆PS₅Cl layer using atomic layer deposition (ALD) (Figure 3e).¹⁹ The physical separation achieved by this approach resulted in remarkable electrochemical cycling performance, with a capacity retention of 92.3% over 400 cycles.

Second, the protective layer does not fully address the mechanical instability of Li metal. As mentioned earlier, high stacking pressures are often employed in halide-based batteries to enhance cathode stability. However, at such pressures, Li metal tends to penetrate the solid electrolyte, even with a more mechanically robust Li-In anode.³⁸ This phenomenon is demonstrated in Figure 3f, where dendrite growth into



Figure 3. Halide electrolyte–anode instabilities. (a) Schematic illustration of (electro)chemical and mechanical instability at the anode–halide interface. (b) Calculated thermodynamic equilibrium voltage profile and phase equilibria of L_3^{1} YCl₆. Reprinted with permission from Reference 7. © 2019 Wiley. (c) Schematic illustration shows the Li/Li₆PS₅Cl/Li₃YCl₆ design (top) and Li/Li₃YCl₆ design (bottom). Reprinted with permission from Reference 36. © 2021 Royal Society of Chemistry. (d) Time-of-flight-secondary ion mass spectrometry images of the interfacial area between Li₃InCl₆ and Li₆PS₅Cl. Shown are secondary ion images of negatively charged fragments S⁻ and InS⁻, indicating the formation of an interphase within the bilayer separator between Li₃InCl₆ and Li₆PS₅Cl. Reprinted with permission from Reference 37. © 2022 Wiley. (e) Schematic representation of assembled cell configuration that consists of depositing a nanometer-thick surface protective layer of Li₃PO₄ made by atomic layer deposition between Li₃InCl₆ and Li₆PS₅Cl. Reprinted with permission from Reference 19. © 2022 American Chemical Society. (f) Scanning electron microscope image of the cell after 100 cycles. The cells were cycled at 3.8 mA cm⁻² under a pressure of 150 MPa within the potential range of 2.1–3.68 V. Reprinted with permission from Reference 29. © 2021 Springer Nature. (g) Schematic of the gradient structural interphase layer generated at the Li/Li_{0.388}Ta_{0.238}La_{0.475}Cl₃ interface. The red double-headed arrows show the Li⁺ flux. Reprinted with permission from Reference 45. © 2023 Springer Nature. (h) Voltage profile of a Li/Li_{0.388}La_{0.475}Cl₃/Li symmetric cell cycled under a current density of 0.2 mA cm⁻² and areal capacity of 1 mAh cm⁻² at 30°C. Insets: corresponding magnified voltage profiles indicate steady Li plating/stripping voltages. Reproduced with permission from Reference 45. Copyright 2023 Springer Nature. SEI, solid-electrolyte interphase; VGCF, vapor-

the electrolyte cannot be completely suppressed when operated under high stack pressure (150 MPa) and high current densities.²⁹

To make halide-based batteries competitive with other solid-state counterparts, researchers are striving to develop Li-compatible halides that can function effectively without the need for a protective layer. Recent studies have focused on modifying halide composition to enlarge ESWs and suppress Li dendrite. Incorporation of Li binary compounds that are ionically conductive and electrochemically stable have shown promise in constructing a stable Li–halide interface.³⁹

Among Li binary compounds, LiF has been regarded as one of most effective SEI components due to its low electronic conductivity and high surface energy (73.28 meV Å⁻²), which inhibits Li dendrite formation.⁴⁰ Moreover, the small lattice constant of LiF allows the SEI to deform elastically with Li metal that constantly changes its morphology.⁴¹ Thus, constructing LiF-rich SEIs has been found to be effective in suppressing Li dendrites and preventing side reactions between halides and Li metal. Inspired by this concept, numerous efforts have been devoted to modulating fluorine-doped halide electrolytes. For example, Li symmetric cells with $Li_3Ybr_{5.7}F_{0.3}$ electrolyte exhibited good stability over 1000 h at 0.75 mA cm⁻² with the capacity of 0.75 mAh cm⁻².⁴² Meanwhile, fluorine doping can also prohibit the reduction of halide electrolytes. A full cell with $Li_2ZrCl_{6-x}F_x$ electrolyte and Li metal showed a 76% capacity retention over 70 cycles due to *in situ* formed F-rich and Cl-rich interphases.⁴³ Nonetheless, it should be noted that fluorine substitution can adversely affect the ionic conductivity of the electrolyte.

The decrease in ionic conductivity caused by fluorine substitution can be partially mitigated by incorporating metal oxide nanoparticles. Jung et al. demonstrated that oxygen substitution at the halide-oxide interface distorts lattice structure and enlarges the Li⁺ transport channel, thereby enhancing Li migration in the electrolyte.⁴⁴ An example of such a halideoxide nanocomposite solid electrolyte is $ZrO_2 - 2Li_2ZrCl_5F$, which exhibits improved Li⁺ conductivity compared to Li_2ZrCl_5F (0.49 versus 0.35 mS cm⁻¹). Importantly, the halide-oxide nanocomposite also demonstrates enhanced highvoltage stability and improved interfacial compatibility with Li_6PS_5Cl as the separating electrolyte.

Recently, a groundbreaking study by Yao et al. introduced a novel LaCl₃-based lithium superionic conductor, Li_{0.388}Ta_{0.238}La_{0.475}Cl₃, which combines Li-metal stability with high ionic conductivity.⁴⁵ Compared to a conventional Li₃MCl₆ lattice, the UCl₃-type LaCl₃ lattice features large, one-dimensional channels for rapid Li⁺ conduction. Through Ta doping and the presence of La vacancies, these channels are interconnected, forming a three-dimensional network for Li⁺ migration. The optimized Li_{0.388}Ta_{0.238}La_{0.475}Cl₃ electrolyte demonstrates an exceptionally high Li⁺ conductivity of $3.02 \,\mathrm{mS \, cm^{-1}}$ at 30°C, coupled with a low activation energy of 0.197 eV. Additionally, as demonstrated in Figure 3g, the novel LaCl₃-based electrolyte generates a gradient passivation layer that efficiently relieves interfacial strain during Li stripping/plating, providing mechanical stabilization for the Li metal electrode. Consequently, the LaCl₃-based electrolyte enables long-term cycling of a Li symmetric cell (1 mAh cm^{-2}) for more than 5000 h (Figure 3h). Theoretical calculations further suggest that metal chlorides with lanthanide family, including the $LnCl_3$ -type chemistries (Ln = La to Lu), exhibit rapid Li⁺ conduction, offering a diverse range of options for halide electrolyte compositions.

In summary, the Li-halide interface encounters challenges such as unfavorable (electro)chemical reactions, nonuniform stripping/plating. and Li dendrite growth, which lead to high interfacial resistance and cell failure. To address these challenges, effective strategies have been identified, including (1) utilizing Li alloys to increase the anode redox potential, thereby reducing reactivity; (2) implementing protection layers at the Li-halides interface to ameliorate interfacial Liion transport and suppress undesirable side reactions; and (3) modifying halide electrolyte composition to improve reductive electrochemical stability and suppress Li dendrite formation. Although each of these methods has its limitations, it is crucial to focus on developing electrolytes that exhibit compatibility with high ionic conductivity, deformability for intimate solid–solid contact, and a wide electrochemical window.

Conclusion and outlook

The past few years have witnessed rapid growth in the variety of halide electrolytes and evolving cell configurations to exploit the full potential of halides. However, interfacial instability remains a critical challenge that needs to be addressed to translate halides' remarkable material properties to excellent cell performance.

On the cathode side, the major challenge lies in mechanical instability caused by limited solid–solid contact and void formation due to volume changes. Although several approaches have been proposed to address this issue, they often come at the cost of compromising electrode energy density or requiring harsh cycling conditions. Halide-based materials are generally considered electrochemically stable at high potentials, but certain materials still experience (electro)chemical side reactions, necessitating time-consuming aging processes to establish a favorable interphase.

On the anode side, conventional halide electrolytes exhibit poor cathodic stability, except for a recent work by Yao et al.⁴⁵ Electronically conductive decomposition products promote continuous side reactions, exacerbating the instability. To address this, either a high-potential Li alloy anode or an additional Li-compatible electrolyte layer can be employed. However, metal anodes are also prone to other mechanical issues such as dendrite growth and void formation. Recent advancements have shown promising possibilities to overcome Li metal incompatibility with dedicated materials development efforts.

Figure 4 illustrates several proposed research directions to tackle these instabilities in halide-based systems. First, mechanical instability in the cathode can be resolved by engineering active materials to negate volume changes and prevent contact loss. Zero-strain cathode materials have recently gained considerable attention due to their potential in solid-state battery applications.^{46,47} These materials offer the advantage of maintaining solid–solid contact between the active material and electrolyte without compromising cutoff voltage, active material fraction, or stack pressure. However, their chemical and electrochemical compatibility with halide electrolytes remains unclear, and research in this area is expected to increase significantly.

Second, particle size and mechanical ductility of the solid electrolyte play crucial roles in achieving intimate solid-solid contact. Downsizing electrolyte particles can enhance cathode active material utilization.⁴⁸ Moreover, smaller particle size helps reduce contact gaps between interfaces and suppresses stress-induced damages in cathode



materials, evidenced by recent findings with <200-nm-sized $\text{Li}_3 \text{InCl}_6$ electrolyte.⁴⁹ Notably, such small particle size is achieved through freeze-drying in aqueous media. The water-compatible processability is a unique advantage of certain halide electrolytes, compared to moisture-sensitive sulfides and oxides that require a subsequent sintering process. Increasing electrolyte ductility is another approach to achieving intimate solid–solid contact. Gallium fluoride-based electrolytes show promise in this regard due to their pliable features and high ionic conductivity.^{50,51}

Third, enhancing electrolyte conductivity is crucial to mitigate the consequences of mechanical instability in the cathode. Higher ionic conductivity compensates for the Li flux hindered by contact loss.⁵² Experimental studies have shown that increasing the cycling temperature enhances ionic conductivity and eliminates the need for high stack pressure to maintain solid–solid contact.¹⁵ The demand for ionic conductivity in electrolytes also arises from the highly tortuous nature of composite cathodes, which leads to longer ion conduction pathways, especially in high-loading and high-rate applications.⁵³ However, there is still a gap between the current state-of-the-art halide electrolytes and conventional liquid electrolytes in terms of achieving optimal conductivity levels.

Fourth, ensuring Li-metal compatibility is critical for solidstate batteries to become competitive in energy density, necessitating the implementation of a Li metal anode.⁵⁴ For halides with poor cathodic stability, using Li metal requires either the addition of an ion-conducting and electron-blocking interlayer or material innovation to form insulative interphase with Li.⁴⁵ While the interlayer approach introduces more interfaces that increase impedance and complicate manufacturing, the use of moisture-sensitive interlayers such as sulfides also undermines the advantage of halide in terms of processability. Therefore, the development of novel Li-stable halides appears to hold promise for practical application in solid-state batteries.

Fifth, achieving interface stability within a reasonable stack pressure range is essential. Excess stack pressure leads to the need for additional packaging devices, ultimately reducing cell-level energy density. Operating cells under low stack pressure, ideally less than 1 MPa,⁵⁵ is desirable for scale-up applications, especially for large-format pouch cells. However, the requirement for low stack pressure exacerbates interfacial instability since voids and cracks can no longer be effectively remedied by mechanical force. The aforementioned strategies offer potential solutions that enable inherently stable interfaces even at low pressure. Nevertheless, there are currently limited reports on the room-temperature and low-stack pressure

operation of halide-based solid-state batteries. Further research is needed in this area to address the challenges associated with interfacial stability under these specific conditions.

Finally, advanced characterization tools play a crucial role in gaining a deeper understanding of interfaces in halide-based batteries. The vulnerability and small scale of interfaces pose challenges for characterization techniques. Chemical instability has been studied using in situ techniques such as XPS and ToF-SIMS during Li plating or surface layer sputtering.^{27,56-58} Ion beam-assisted cross-section analysis and x-ray tomography are employed to characterize morphological instabilities such as particle crack, void formation, and contact loss.^{10,59,60} While extensive interface studies have been reported for oxides and sulfides, we anticipate a significant increase in studying halide interface utilizing these techniques in the near future. These advanced characterization methods will enable a comprehensive understanding of halide-based battery interfaces and accelerate the development of effective strategies to enhance their stability and performance.

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Data availability

Not applicable.

Code availability

Not applicable.

Conflict of interest

Y.Y. has equity interest in LiBeyond, LLC and Solid Design Instruments, LLC. The University of Houston reviewed and approved their relationship in compliance with its conflict-ofinterest policy. The remaining authors declare no competing interests.

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