

Toward Practical All-solid-state Batteries with Sulfide Electrolyte: A Review

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Abstract Sulfide-based solid-state electrolytes with ultrahigh lithium ion conductivities have been considered as the most promising electrolyte system to enable practical all-solid-state batteries. However, the practical applications of the sulfide-based all-solid-state batteries are hindered by severe interfacial issues as well as large-scale material preparation and battery fabrication problems. Liquid-involved interfacial treatments and preparation processes compatible with current battery manufacturing capable of improving electrode/electrolyte interface contacts and realizing the mass production of sulfide electrolytes and the scalable fabrication of sulfide-based battery component have attracted considerable attention. In this perspective, the current advances in liquid-involved treatments and processes in sulfide-based all-solid-state batteries are summarized. Then relative chemical mechanisms and existing challenges are included. Finally, future guidance is also proposed for sulfide-based batteries. Focusing on the sulfide-based all-solid-state batteries, we aim at providing a fresh insight on understandings towards liquid-involved processes and promoting the development of all-solid-state batteries with higher energy density and better safety.

Keywords Sulfide electrolyte; All-solid-state battery; Composite electrode; Liquid-involved materials processing; Energy material

1 Introduction

Since the successful fabrication of lithium ion batteries in 1991, advanced electrochemical energy storage technologies have been integrated into every aspect of our lives and have completely promoted our daily lives^[1–9]. However, the energy density of commercialized lithium-ion batteries is approaching their theoretical limit, which is gradually falling behind the demands of rapidly developing portable electronic devices and electric vehicles for higher energy density^[10–19]. Furthermore, with the increase of energy density, the adaption of flammable organic liquid electrolytes causes severe safety concerns.

All-solid-state batteries based on inorganic solid electrolyte are considered as the ultimate solution to achieve both high energy density and excellent safety^[20–30]. On the one hand, the wide electrochemical window of solid-state electrolyte(SSE) makes it easily compatible with lithium metal anode and high voltage cathodes, therefore, significantly improving the battery system energy density. More importantly, using SSE to replace

routine liquid electrolyte can avoid the leakage of liquid electrolyte and mitigate the risk of combustion accident and even explosion.

Sulfide-based SSEs, such as $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS)^[31–33], $\text{Li}_{9.54}\text{Si}_{1.74}\text{P}_{1.44}\text{S}_{11.7}\text{Cl}_{0.3}$ ^[34,35] and $\text{Li}_6\text{PS}_3\text{Cl}$ ^[36–38], possessing high lithium ion conductivity of more than 10^{-3} and even 10^{-2} S/cm have attracted considerable attention and are widely regarded as the most promising electrolyte system to enable the practical applications of all-solid-state batteries^[39–47]. Unfortunately, all-solid-state batteries suffer from a huge challenge in charge transport at electrode/electrolyte interface^[43,48–53]. Unlike liquid electrolyte enabling smooth interfacial charge transport in a conventional battery, limited solid-solid contact between electrode and electrolyte in a solid-state battery leads to the increased interface impedance and inferior charge transport capability^[54–62]. In addition, sulfide electrolyte is moisture-sensitive, which can react with water to form poisonous gas^[63–65]. In view of practical application of solid-state batteries, concerns on health problems during relative processes of

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electrolyte synthesis and battery fabrication should be considered.

Recently, liquid-involved strategies, including interfacial therapy of liquid solution, wet-chemical synthesis of sulfide electrolyte and solution-processable battery fabrication have drawn numerous attentions in sulfide-based all-solid-state batteries^[66–68]. In view of paramount effects of interfacial charge transfer on solid-state batteries, liquid-phase therapy that is using the mobility and permeability of additional liquid precursors to construct conformed electrode-electrolyte interface, is regarded as an effective strategy to reduce interfacial ion transport resistance^[69,70]. In comparison to solid-phase synthesis at elevated temperature, liquid-phase syntheses of sulfide electrolytes exhibit distinct advantages in low cost, low time consumption, and scalable production. Distinguishing from solid-state reaction from dry ingredients at small scale, solution-involved processes are also more appropriate for large-scale fabrication of solid-state electrolyte and solid-state electrode layers^[71–73]. Therefore, liquid-involved processes are of great significance with respect to not only the fabrication of sulfide electrolytes and solid electrodes, but also the high-efficient operation of sulfide-based all-solid-state batteries.

In this perspective, we summarized the recent advances of liquid-involved processes in interface improvement, material preparation, and manufacturing process. Meanwhile, theoretical views and chemical/electrochemical mechanisms are discussed. Finally, existing challenges and future directions are also out looked.

2 Liquid Wetting of Solid Electrode/Electrolyte Interface

In solid-state batteries, the poor solid-solid interfacial contact between electrolyte and electrode severely limits Li ion

transport and introduces large interfacial impedance, deteriorating the electrochemical performance. In this regard, the construction of a conformal contact interface by interfacial wetting with a liquid solution is considered as one of the most effective and convenient strategy^[66]. The high mobility of liquid solution not only contributes to an increased contact area, but also effectively facilitates the ion migration capability. Organic liquid electrolytes, such as carbonate electrolyte, are the most frequently adopted wetting reagents for SSEs, especially for oxide-based SSEs; yet it is rarely operated in sulfide-based solid-state batteries due to the high dissolution of sulfide materials into organic polar solvents. Distinguish from the routine liquid electrolyte, the highly concentrated electrolyte with low amount of free solvent molecules helps to alleviate the incompatible problems. Gewirth *et al.*^[74] employed high-concentration electrolyte (MeCN)₂-LiTFSI:TTE as a wetting interlayer between electrodes and sulfide SSE, which enabled highly intimate and stable contact interfaces so that it could afford favorable Li ion transfer pathway, thereby promoting the ion transport kinetics [Fig. 1(A)].

Besides, room temperature ionic liquid (IL) constitutes another potential candidate for interfacial wetting in sulfide electrolytes, attributing to its high ion conductivity, wide electrochemical window, excellent thermal stability, less volatility, nonflammability, and superior compatibility against sulfide SSEs^[75]. Jung and co-workers^[76] implemented Li(G3)TFSI (triethylene glycol dimethyl ether, G3) IL into the sulfide solid state batteries to enhance the interfacial wettability. With the addition of LiG3, the contact modes were converted from imperfect solid-solid contacts to favorable ionic solid-liquid-solid contacts, rendering the solid-state batteries containing LFP cathode with a high capacity performance of 144 mA·h/g, which was dramatically contrasted by the negligible capacity without LiG3 [Fig. 1(B, C)].

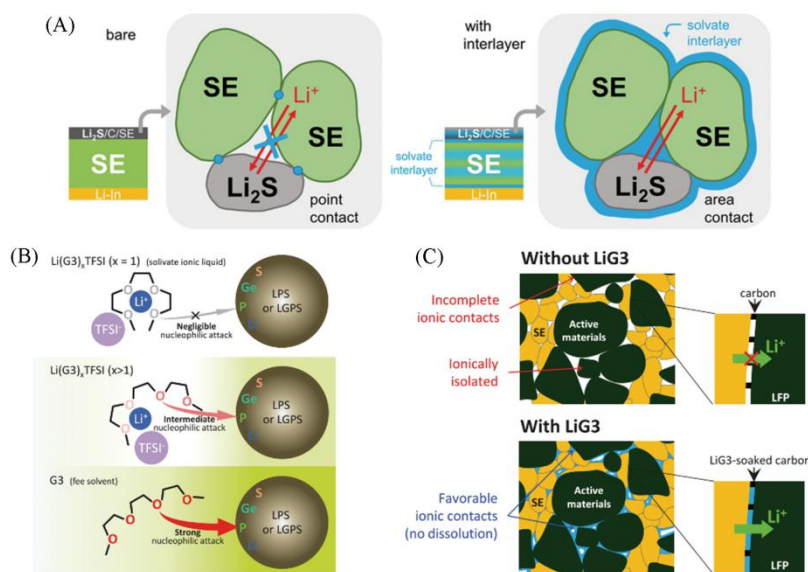


Fig.1 Liquid-phase infiltration and wetting strategy for solid-solid interface in cathode

(A) Schematic diagram of the solid-state Li₂S batteries with high-concentration electrolyte solvate interlayer(right) and without liquid-phase wetting(left); (B) schematic of the reactivity of sulfides with different glyme-based liquids; (C) the change of electrode macrostructure with/without solvated Li(G3)TFSI ionic liquids.

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Noteworthy, the interfacial instability is another critical issue affecting battery performance in addition to inferior contact for the Li metal-involved interfaces^[77–83]. Due to the low potential of Li metal, the sulfide electrolyte can be easily reduced and forms a low ionic conductive layer between sulfide SSE and Li anode interface^[84–90]. The formation of electronically conductive Li-Ge alloy further leads to gradual degradation of electrolyte and propagation of interfacial layers especially for LGPS SSE, resulting in rising resistance and rapid capacity decay, and seriously short-circuiting of batteries^[91,92].

In order to stabilize the sulfide SSE/Li anode interfaces, the strategy of *in-situ* formed solid electrolyte interphase layer(SEI) by the reaction of metallic lithium with liquid wetting agent along with interface therapy process synchronously is effective. Yang and co-workers^[84] added *N*-methyl-

N-propylpyrrolidinium bis(trifluoromethanesulfonyl)imide (PYR₁₃TFSI) ionic liquid containing lithium bis-(trifluoromethanesulfonyl)imide(LiTFSI) to the LGPS surface, which worked as the interface modifier and contributed to the formation of *in situ* SEI layer, resulting in high interface stability. Consequently, stable Li deposition over 1200 h lifespan with a small interfacial resistance was achieved for symmetric Li/Li cell(Fig.2). By regulating the interfacial reactions, both composition and structure of SEI layer can be manipulated, thus impacting the electrochemical performance. Zheng *et al.*^[85] compared the interfacial behavior of Li salt/PYR₁₃TFSI, and proposed the presence of LiFSI salt leads to LiF enrichment in SEI layers, bring about higher interface resistance and poor cycling performance.

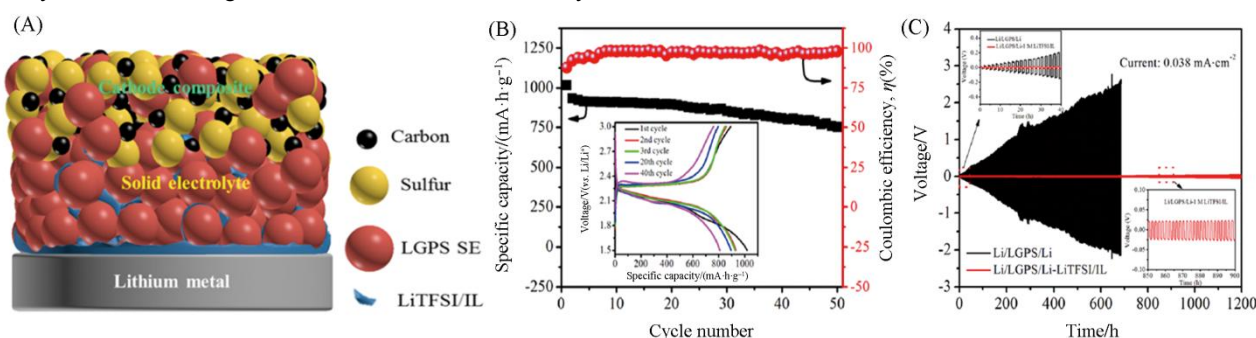


Fig.2 Liquid-phase therapy for metallic Li/sulfide electrode interface

(A) Schematic representation of protected lithium anode by *in situ* formed solid electrolyte interphase layer in a solid-state lithium-sulfur battery; cycling performances(B) and anodic interface stability(C) of solid-state lithium-sulfur batteries.

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The liquid-phase wetting strategy is helpful to construct a conformal interface between sulfide SSE and electrode to contribute intimate contact, which affords enough Li ion transport channels, effectively reducing interfacial resistance and improving cycling performance. Moreover, by taking advantage of the interfacial reactions between liquid wetting reagents and Li metal, it is also profitable for constructing protective layers at the sulfide electrolyte/Li interface to prevent the continuous reduction of sulfide SSEs by Li metal, ensuring excellent interfacial stability. However, the introduction of liquid substances significantly changes the electrode/electrolyte interfaces from single solid-solid interface to two solid-liquid and liquid-solid interfaces, which brings about sophisticated interfacial behaviors accompanied with severe side reactions. During repeated cycling, these unexpected reactions not only reduce the utilization of active materials, but aggravate the consumption of limited wetting reagent, leading to subsequent interface failure and eventually interfacial ion transport decay. Therefore, clear understanding of the interfacial ion transport and interfacial (electro)chemical behaviors at conformed solid-liquid interface is important for extending liquid-phase therapy strategy in sulfide-based solid-state batteries.

3 Liquid-involved Preparation of Sulfide Electrolyte

In general, the sulfide-based SSEs are prepared by high-energy ball milling and high-temperature annealing,

which are energy-intensive and time-consuming processes and thus hinder the large-scale application of sulfide electrolytes in battery manufacturing^[35]. In 2013, β -Li₃PS₄ electrolyte was synthesized by Liang and coworkers^[93] via a simple and feasible liquid-phase chemical reaction between Li₂S and P₂S₅ in tetrahydrofuran(THF) solvent[Fig.3(A, B)]. After that, a series of sulfide electrolyte including Li₇P₃S₁₁ and Li₆PS₅X(X=Cl, Br, I) has been prepared by this liquid-phase chemical reaction between Li₂S and P₂S₅ precursors as well as other additives, such as LiCl, LiI and so on^[72,94–97]. Surprisingly, the liquid-phase synthesis also provides the opportunities to explore new sulfides electrolytes, such as Li₇P₂S₈I and Li₄PS₄^[98–101].

In addition, another liquid-involved dissolution-precipitation method, in which the pre-prepared sulfides are firstly dissolved into organic solvent and then the sulfide SSEs are re-obtained by their recrystallization, reprecipitation or solution evaporation has also been reported to synthesize sulfide-based electrolyte. The particle size, microstructures and surface morphologies of SSE can be controllably reconstructed by the recrystallization and reprecipitation^[102,103]. For instance, uniform and submicrometric-sized Li₆PS₅Cl particles were obtained by the reprecipitation of sulfide electrolyte^[104]. Flake-like Li₃PS₄ electrolytes were also controlled by Liang and coworkers^[105,106]. In a solid-state battery, sulfide electrolytes with high specific area favor the interface contacts between electrolyte particles and active materials. The

re-precipitation can be utilized to construct integrated electrode structure with good three-phase interface among the SSE, active materials and the electronic conductors^[107]. In particular, conformed surfaces between electrode materials and electrolytes can be achieved by the evaporation of organic

solvent from sulfide solution and *in-situ* precipitation of dissolved sulfide electrolyte onto the surface of electrode materials, thus significantly promoting the solid-solid contact [Fig.3(C)]^[108–110].

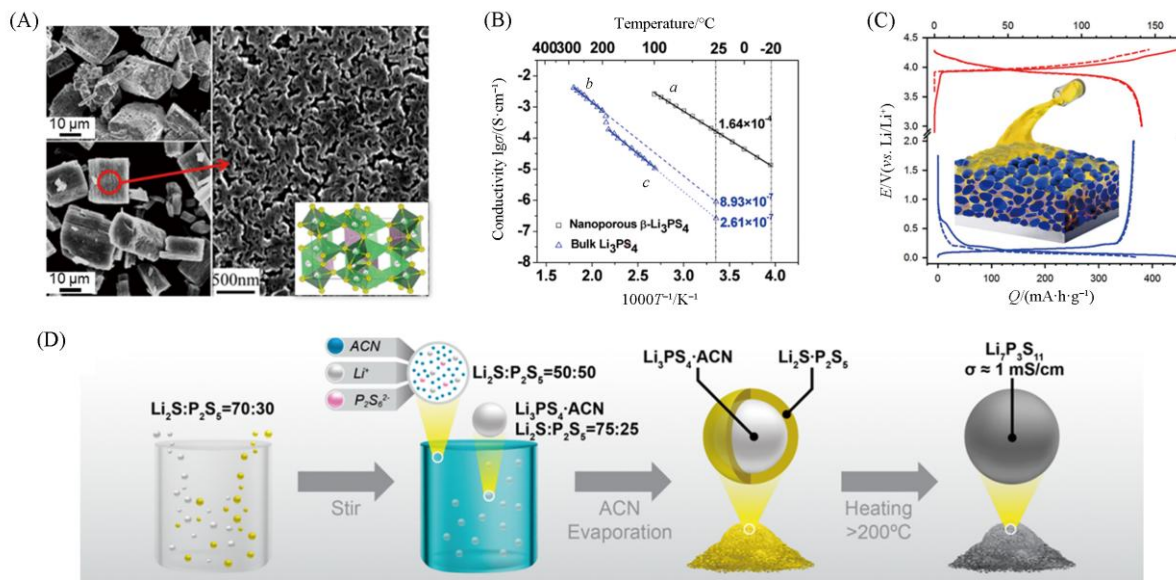


Fig.3 Liquid-involved preparation of sulfide electrolyte

Structure and morphology(A) and ion conductivity(B) of the as-synthesized $\text{Li}_3\text{PS}_4\text{-3THF}$ electrolytes by liquid-phase wet-chemical approach; (C) conformed electrode/electrolyte interface by the infiltration of solution-processable sulfide electrolyte solution into porous electrode; (D) schematic illustration of the reaction mechanism of $\text{Li}_7\text{P}_3\text{S}_{11}$ in acetonitrile solvent.

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There are many superiorities of wet chemical methods in sulfide electrolyte production. However, the sulfide electrolytes by wet chemical processes generally possess a relatively low lithium ion conductivity at room temperature. Wang *et al.*^[97] systematically investigated the reaction mechanism of $\text{Li}_7\text{P}_3\text{S}_{11}$ in an acetonitrile solvent[Fig.3(D)]. The liquid-phase synthesis of sulfide involves three complicated processes: solution chemical reaction, intermediate precursor formation, and crystallization. Therefore, technical parameters(reaction time, reaction temperature, post-treatment temperature, etc.) and related details(solvent type, solvent polarity, as well as the remove of solvents) play vital roles in the formation and precipitation of sulfide electrolyte. Unfortunately, incomplete and inadequate chemical reactions as well as impure residues are adverse to acquire sulfide SSEs of high purity and crystalline phase of high ion conductivity. Actually, solution chemical reactions and precipitation processes are dominated by both the thermodynamics formation and kinetics balances. In-depth understanding of liquid-phase syntheses of sulfide electrolytes should be further conducted.

4 Slurry-processable Fabrication of Sulfide-based Solid-state Batteries

At present, the studies on solid-state batteries are mainly conducted in a mould cell with limited sizes, where the electrolyte layers and solid composite cathodes are almost prepared by powder die compression. Although conventional pressing processes contributed to densifying electrolyte and electrode

layers as well as intimate electrolyte/electrode contacts^[111], it is difficult for traditional pressing technology to obtain a thin SSE layer, which is a guarantee to achieve high energy density. Moreover, this technology is also inappropriate for large-scale manufacturing. Towards the practical applications, solid-state batteries including electrolyte and electrode components should be achieved by scalable industrial processes.

Slurry-coating process is a typical industrial fabrication process and widely used in commercialized lithium ion battery manufacturing. Recently, sheet-type SSE membranes were successfully achieved by solution slurry-coating processes. Sakuda *et al.*^[112] reported a self-standing SSE sheet with a large size of 22 mm×22 mm by coating the sulfide electrolyte slurry on commercialized copper foil *via* a simply slurry coating process, in which the slurry was prepared by mixing sulfide electrolyte, polymer binder, and organic solvent. More importantly, all-solid-state sheet-type batteries assembled by this self-standing SSE sheets exhibited an energy density of over 155 W·h/kg in despite of excluding the mass of current collectors and package. Therefore, this sheet-type structure design attributed to controllable slurry-coating technology shows a huge possibility in achieving high battery energy density. Jung and coworkers^[113] firstly demonstrated a bendable sulfide SSE films with a thin thickness of *ca.* 70 μm, which was prepared through coating electrolyte powder dispersion on porous poly(paraphenylene terephthalamide) nonwoven scaffold using the doctor blade method[Fig.4(A) and (B)]. As a result, the all-solid-state lithium battery fabricated with this thin

electrolyte film showed a 3-fold increase in battery energy density compared with a battery with a conventional powder electrolyte sheet.

For the successful commercialization of solid-state batteries, a feasible and scalable production procedure of composite cathodes is also indispensable. Recently, Passerini and coworkers^[71] developed a composite solid cathode by tape-casting a mixture slurry of binder, conductive carbon, β - Li_3PS_4 and $\text{Li}_{1+x}[\text{Ni}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}]_{1-x}\text{O}_2$ on Al-foil. Meanwhile, the SSE layer was also prepared by the same slurry-coating process. In order to simplify operation process, a two-in-one cathode-supported electrolyte layer was obtained by directly coating an electrolyte slurry on a prepared solid cathode^[112]. Jung and coworkers^[114] further simplified the $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ (NCM) electrode fabrication by a one-pot slurry prepared from SE precursors (Li_2S and P_2S_5). This procedure integrated the pre-preparation of sulfide electrolyte into one-step cathode

production, exhibiting a huge potential in the application of practical solid-state batteries [Fig.4(C)]. All-solid-state lithium-sulfur batteries with higher theoretical energy density have attracted considerable attentions^[115–117]. However, the mass production of solid-state sulfur cathode remains huge challenges^[118–120]. In most of organic solvents, sulfide electrolyte can react with elemental sulfur and thus form soluble polysulfide intermediates. Recently, Zhang and coworkers^[121] developed a large-scale fabrication of sulfur cathode by slurry-coating procedure [Fig.4(D)]. The chemical compatibility between sulfur and sulfide electrolyte was realized by the screening of dielectric constant and polarity of dispersion solvents. The solid sulfur cathode films with tunable thickness and flexibility were prepared by slurry-coating procedure [Fig.4(E)]. As a result, all-solid-state Li-S pouch cells with area capacity of over $2.3 \text{ mA}\cdot\text{h}/\text{cm}^2$ have been realized and the related protocols are also capable of scaled-up manufacturing.

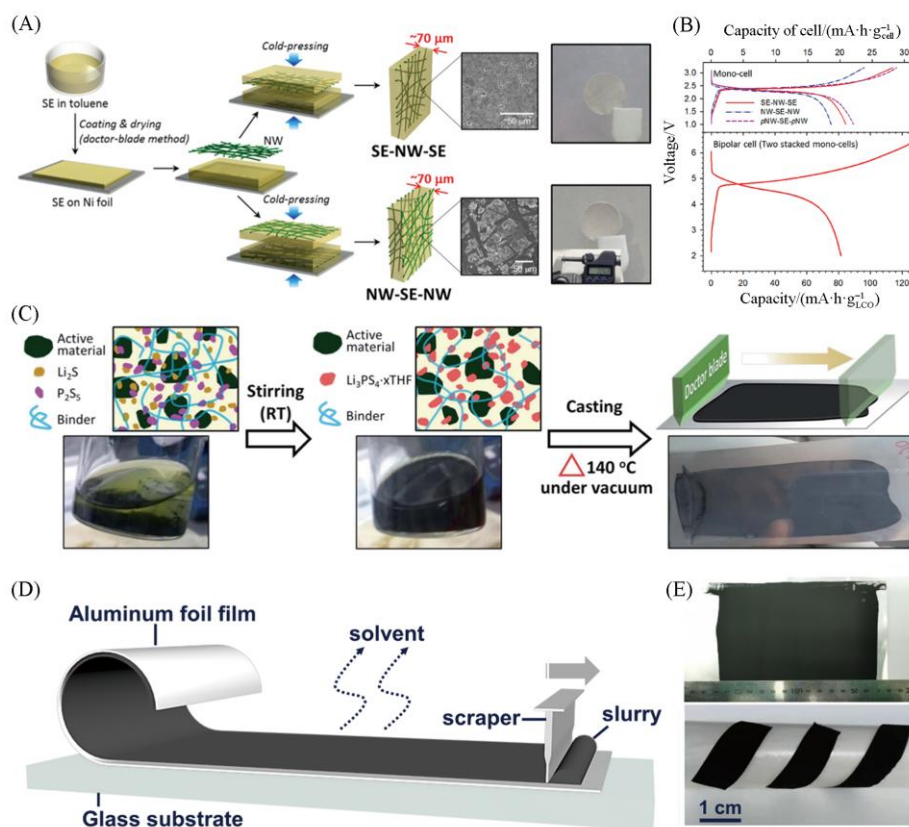


Fig.4 Slurry-processable fabrication of sulfide-based solid-state batteries

(A) Schematic illustration of the fabrication of bendable and thin sulfide solid electrolyte film and their optical photographs; (B) electrochemical performances of the all-solid-state LCO/LTO cells (up) with different-structured electrolyte films and the cell (down) with stackable bipolar structure configuration; (C) schematic diagram of the fabrication of sheet-type composite electrode by a single-step wet-chemical route *via* a tape-casting process, in which the slurry is prepared directly from sulfide SSEs precursors; (D) schematic illustration of the fabrication of solid-state sulfide/sulfur cathode by a slurry-coating process; (E) optical photographs of the as-prepared electrode films.

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Generally, towards electrode fabrication by a slurry-coating process, polymer binder is indispensable, which not only thickens the electrode slurry and regulates its viscosity but also maintains the intimate contact between electrode materials and binds the electrode materials on conductive substrate^[122,123]. However, inactive polymer binders reduce the energy density

of a battery. Recently, Yamamoto *et al.*^[124] firstly reported a binder-free sheet-type battery. The volatile poly(propylene carbonate)-based binders are proposed for SSE batteries. After coating electrolyte slurry on current collectors, the volatilizable polymer binders can be removed during the heat treatment and drying process of sheet-type electrodes (Fig.5). In addition, due

to the absence of inactive binders, charge transport resistances in solid electrodes were also reduced. The solid-state batteries with this electrode sheets exhibited enhanced rate performance

and cyclability. Moreover, the energy density of these batteries also increased by 2.6 times.

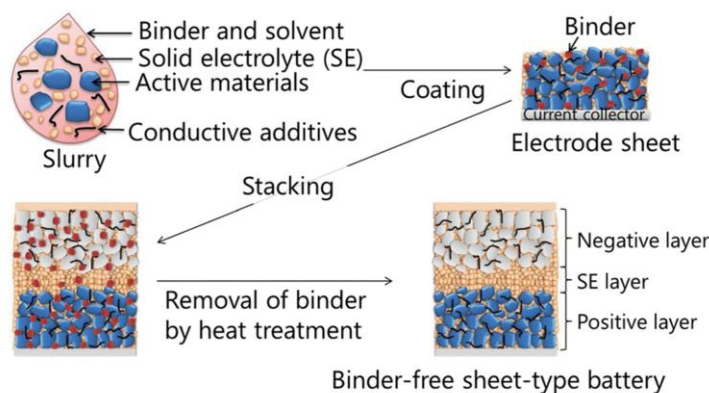


Fig.5 Schematic illustration of the fabrication of binder-free sheet-type all-solid-state battery achieved by the volatily remove of polymer binders under heat treatment process

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To date, slurry-processable preparation technology is regarded as one of the most promising approaches for large-scale fabrication of SSEs and electrodes. Although great breakthroughs have been achieved by scalable battery preparations, complicated issues remain. Firstly, after liquid-involved processes, the guarantees towards the crystallinity and purity of high ion conductivity of sulfide electrolytes are the key issues^[125,126]. The electrolyte layer is mainly responsible for the ion migration between anode and cathode in a solid-state battery. While electrochemical reactions are proceeded in electrode completely, where involves not only ion transport but also electron transfer. Considering the influences of the dissolution and possible chemical reaction of sulfides in a slurry solution on charge transport in solid electrode, how to achieve the compatibility between conductive carbons, ionic conductive sulfides, active materials, polymer binders and dispersion solvent, as well as the uniform distribution of every component in electrode needs to be considered deeply. In addition, utilizing solution-based methods to realize the integral construction between SSEs and electrode is helpful to simplify the battery fabrication with intimate interfacial contact. Therefore, the solution-based processes should be highlighted in future research.

5 Conclusions and Perspectives

Since the research and development of ultrahigh lithium ion conductive sulfide SSEs, advanced lithium batteries based on sulfide SSEs provide considerable chances in achieving high energy density and high safety. However, the practical applications of all-solid-state batteries are limited by several obstacles, such as the mass production of solid sulfide electrolyte materials, the scalable fabrication of battery electrodes, as well as severe interface issues between solid electrolytes and electrodes.

In this perspective, on account of the success in promoting the proceeding of sulfide-based all-solid-state lithium batteries, we specifically highlighted the liquid-involved processes. As we know, limited solid-solid contacts between solid electrode

and electrolyte always lead to inferior ion transport and thus hinder the development of all-solid-state batteries. The liquid-phase wetting approaches have achieved huge successes in improving solid-solid interface contact. Attributing to the fluidity and infiltrability of liquid electrolyte or other liquid-phase solution, the interfaces with lost solid-solid contact can be reconstructed and alternated by new solid-liquid interfaces and thus, significantly reducing the interfacial resistance of ion transport. The liquid-phase synthesis processes of sulfide electrolytes pose significant advantages in the limited energy consumption, reduced time costs, and good operability for industrial preparation of solid sulfide electrolytes. Moreover, this method also provides the possibilities in exploring and developing new type sulfide electrolytes. At the same time, the structure and surface morphology of sulfide electrolytes can be also reconstructed by this liquid-involved method. Notably, conformed electrolyte/electrode surfaces with good solid-solid contact can be achieved by *in situ* coating solid sulfides on electrode materials using a dissolution-precipitation process. In view of practical application of solid-state batteries, the fabrication of electrolytes and electrodes with liquid-involved slurry-coating processes is introduced due to the film-processing ability of this slurry approach to get thin and extended electrolyte and electrode layers. The integrated electrode-electrolyte preparation matched with industrial manufacturing is also developed, which is also capable of obtaining a thinner electrolyte layer for higher energy density.

Although the liquid-involved processes have expedited the proceeding of sulfide-based all-solid-state lithium batteries, there still remain huge challenges. Future contributions should be focused on understandings of liquid-phase reaction mechanisms, interfacial chemical/electrochemical behaviors and engineering fabrication. The corresponding perspectives are also provided as follows:

(1) Interfacial behaviors. Because of the transformation from solid-solid interface into solid-liquid-solid interface after liquid-phase therapy, the interfacial chemical/electrochemical behaviors are also converted synchronously. How about the

interfacial ion transport along with interfacial behavior conversion? In addition, owing to the presence of liquid-phase solution, whether the possible incompatibilities, such as the dissolution of sulfide electrolytes into liquid solution, the reactions between electrode and solution as well as the interphase formation, have an effect on the interfacial stability.

(2) Chemical/electrochemical stability. Li metal possessing the most negative electrochemical potential (−3.040 V vs. standard hydrogen electrode) and high specific capacity (3860 mA·h/g) has been regarded as the most promising anode to achieve enhanced energy density^[127]. However, the interface reactions between Li metal and sulfide electrolytes always impede Li ion migration and result in enhanced interfacial resistance. Moreover, the parasitic side reactions also lead to lithium dendrite formation, causing battery short circuit and even safety hazards. In addition, interface compatibilities of sulfide electrolytes when they were implanted into oxide cathodes also cause a huge challenge in interfacial ion conduction due to the inferior oxidation stability and the space charge layer. Consequently, the chemical and electrochemical stability of sulfide electrolytes towards Li metal anode as well as high voltage oxide materials needs to be further improved.

(3) Evolution of solid-solid interface. To date, the issues underlying solid-solid interfaces are still the main limiting factors for the development of solid-state batteries^[128]. The initial solid-solid contacts can be integrated well under static pressure. However, a stable electrochemical performance of a solid-state battery is difficult to be obtained, which is generally attributed to the degradation of electrode/electrolyte interfaces. Therefore, the evolution of solid-solid interfaces should be investigated during operating.

(4) Sulfide liquid-phase formation mechanism. Although wet-chemical synthesis routes have achieved the production of sulfides and even the new sulfide varieties, the liquid-phase reaction mechanism is still elusive. Generally, the sulfide electrolytes prepared by liquid-phase processes show low crystallinity and inferior purity and thus, rendering low ionic conductivity. Therefore, understanding on the liquid-phase formation mechanisms is imperative and essential for further realizing sulfide preparation with high ion conductivity. Moreover, the compatibility against solvent, active materials and binders in liquid-phase solution should also be considered.

(5) Environmental compatibility. The liquid-phase synthesis and solution-involved process of sulfide SSEs and electrode generally need to use toxic solvents. Moreover, conventional sulfide electrolytes are usually unstable in moisture atmosphere and can react with water to generate toxic H₂S gas, leading to severe health and environmental concerns. Therefore, air-stable sulfide electrolytes and more environmentally compatible preparation approach should be developed. In addition, the reuse and recovery of relative solvents needs to be also considered.

Overall, despite the achievements using liquid-phase processes in sulfide-based all-solid-state batteries, there still remains a huge open space. Much more efforts deserve to be devoted to promoting the development of practical all-solid-state batteries with the combination between fundamental

chemical/electrochemical understandings, advanced characterization technologies and engineering researches in the future.

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