

Mini Review

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High-stability room temperature ionic liquids: enabling efficient charge transfer in solid-state batteries by minimizing interfacial resistance

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How to cite this article: Jeong S, Ho VC, Kwon O, Park Y, Mun J. High-stability room temperature ionic liquids: enabling efficient charge transfer in solid-state batteries by minimizing interfacial resistance. *Energy Mater* 2023;3:300048. <https://dx.doi.org/10.20517/energymater.2023.47>

Received: 9 Jun 2023 **First Decision:** 10 Jul 2023 **Revised:** 28 Jul 2023 **Accepted:** 1 Sep 2023 **Published:** 2 Nov 2023

Academic Editors: Giovanni Battista Appetecchi, Yuping Wu **Copy Editor:** Fangyuan Liu **Production Editor:** Fangyuan Liu

Abstract

Currently, intensive research is underway to develop stable electrolyte systems that can significantly enhance the performance of rechargeable batteries. Recent advances in solid electrolytes have led to new types of promising systems owing to their high conductivity. This has generated considerable interest in the practical applications of safe batteries. Considering the safety concerns associated with rechargeable batteries, solid electrolytes have become indispensable for the advancement of next-generation battery technologies. However, the increased interfacial resistance at solid-solid interfaces has become a critical challenge. To address this problem, room-temperature ionic liquids (RTILs) have been investigated as functional materials for mitigating the interfacial resistance in solid-state batteries (SSBs). The special properties of RTILs, such as their non-volatility, non-flammability, and high safety characteristics, make them highly promising candidates for safe batteries. Various approaches have been explored for the effective utilization of ionic liquids in SSBs. This review provides a comprehensive discussion on the application of RTILs as electrolytes, considering their electrochemical properties and incorporation into composites to minimize resistance in SSBs.

Keywords: Room temperature ionic liquid, room temperature molten salt, all-solid battery, solid-liquid interface, solid electrolyte



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INTRODUCTION

The importance of rechargeable battery systems in addressing and mitigating global issues, such as climate change and global warming, is increasing. Over the past decade, the rechargeable battery market, led by electric vehicles and large-scale energy storage systems, has experienced rapid growth. In particular, lithium-ion batteries (LIBs) have been widely used as optimal secondary battery systems for energy conversion and storage since their initial commercialization in 1991. LIBs must satisfy various performance criteria, including energy, power, cyclability, safety, cost, environmental friendliness, and temperature ranges. Considering these parameters, LIBs are the most widely utilized electrochemical energy-conversion devices and provide the highest level of performance.

With the recent emergence of numerous applications requiring kWh-to-MWh-scale secondary batteries, safety has garnered significant attention. Recently, several fire accidents involving electric vehicles and energy storage systems have been reported^[1]. Considering the history of the downfall of the lithium metal battery market owing to safety concerns regarding lithium dendrite failures, addressing the safety issues of secondary batteries is one of the most critical challenges in their applications^[2-4]. The key components of secondary batteries are the cathode, anode, electrolyte, and separator. For conventional LIBs, carbonate-based electrolytes are utilized to provide a robust solid electrolyte interface (SEI) on the negative electrolyte and the high solubility of lithium salts. From a safety viewpoint, carbonate-based electrolytes pose inherent challenges owing to their volatility, low flashpoints, and high flammability. However, these electrolytes offer high battery performance through excellent film formation and wetting characteristics on graphite, making it difficult to transition to safer alternative electrolytes.

Room-temperature ionic liquids (RTILs) are unique solutions that exhibit both ionic and liquid states, even at room temperature^[5-8]. The distinctive characteristics of RTILs are that they are non-volatile, with negligible ignition points and flammability, making them excellent and safe electrolyte candidates for LIBs. Considering the flammability issues often associated with liquid electrolytes, RTILs have great safety potential for applications in LIBs. Although attempts have been made to use RTILs as electrolytes for lithium secondary batteries for a long time, their application has been limited because of their electrochemical reactivity, low conductivity, and corrosiveness to electrode materials^[9-12].

There has been a growing interest in the development of solid electrolytes for high-safety solid-state batteries (SSBs). Solid electrolytes in batteries not only possess excellent thermal stability but also prevent the rapid spread of battery ignition by minimizing electrolyte leakage, even if the battery is damaged^[13,14]. $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS), which has a conductivity comparable to that of liquid-type carbonate electrolytes, has been perceived to surpass the typical low-ion conductivity limits of solid electrolytes^[15]. However, the fabrication and operational processes of conventional LIB systems have been developed based on liquid electrolytes, which has led to the identification of numerous performance limitations and challenges for SSB systems. Therefore, new electrochemical systems are currently being investigated to address these issues through the introduction of electrode designs and functional materials^[16,17].

In particular, the interfacial resistance of SSBs is considerably high because of their exclusively solid interfaces, unlike routine electrochemical systems with liquid electrolytes [Figure 1]^[18-20]. In batteries, electrochemical reactions occur at the interfaces where ions and electrons can be transported simultaneously. Liquid electrolytes act primarily as ion conductors, whereas carbon powder and metal substrates serve as electronic conductors. Finding electrode systems that demonstrate excellent ion and electron conductivities in SSBs poses a significant challenge to achieving high-performance SSBs. The formation of ternary interfaces among the active materials, conductive carbon powder (as the electrode

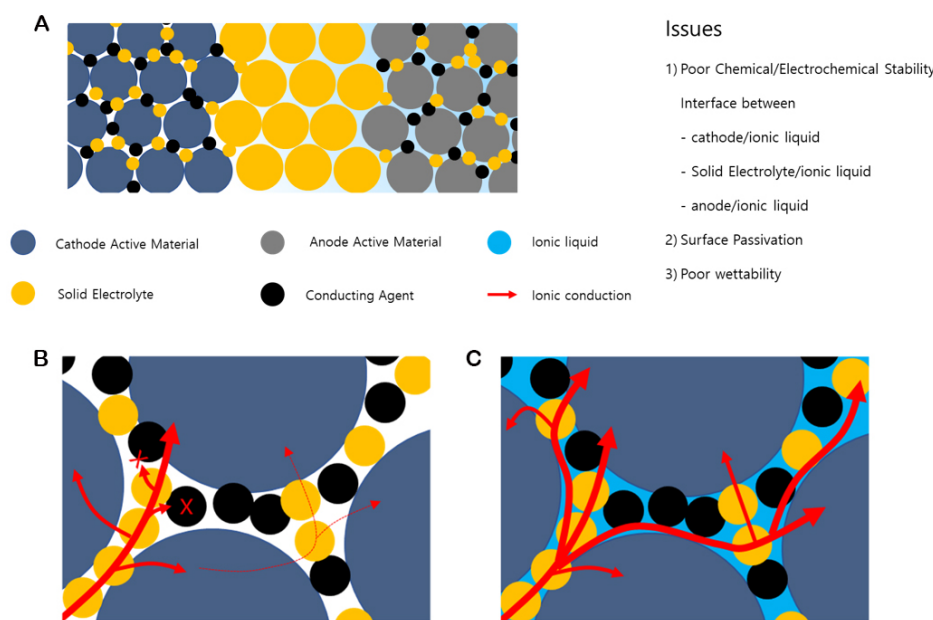


Figure 1. (A) Schematic figure of all-solid batteries with active materials (cathodes and anodes), conducting agents, solid electrolytes, and ionic liquids. Enlarged particle interface among conducting agents, solid electrolytes, and active materials (B) without ionic liquids or (C) with ionic liquids.

conducting agent), and solid electrolytes with high ionic conductivities is limited. Owing to this limitation, solid electrolytes have a less electrochemically active interface than liquid electrolytes, resulting in a high interfacial resistance^[21].

Incorporating a small amount of RTILs as a supporting liquid electrolyte in SSBs is highly effective. The integration of a small number of nonflammable RTILs into SSBs aligns effectively with the goal of enhancing battery safety and contributing to the reinforcement of battery safety issues. The non-volatile characteristics of RTILs match the merits of SSBs. In this review, we summarize the studies on the use of RTILs in SSBs. Additionally, we discuss the phenomenological issues related to the reduction in interfacial resistance highlighted in the relevant literature and explore future directions.

ELECTROCHEMICAL STABILITY OF IONIC LIQUID

Ionic materials typically exist in the solid state at room temperature owing to the strong coulombic interactions between cations and anions. Similarly, RTILs primarily consist of cationic and anionic molecular structures, the structural stability and properties of which are generally determined by large structures to delocalize Coulombic interactions. These bulky ionic molecular structures allow the introduction of various substituents, so they are frequently referred to as designer solvents with various characteristics. Various forms of RTILs combining different types of cations and anions have been proposed. Suitable electrolytes for electrochemical systems and rechargeable batteries need to possess essential electrochemical properties such as high electrochemical stability and ionic conductivity. In the early stages of RTILs, aluminum-chloride-based ions were used as anion sources; however, they exhibited poor moisture stability and high metal corrosiveness, making them unsuitable as electrolytes for secondary batteries^[22,23]. As shown in Figure 2, the RTILs were prepared with bulky anions, which are the same as the lithium salts in the liquid electrolytes used for LIBs. Imidazolium, pyrrolidinium, piperidinium, and quaternary ammonium are used as cations, whereas anions, such as TFSI and FSI, from the fluorinated

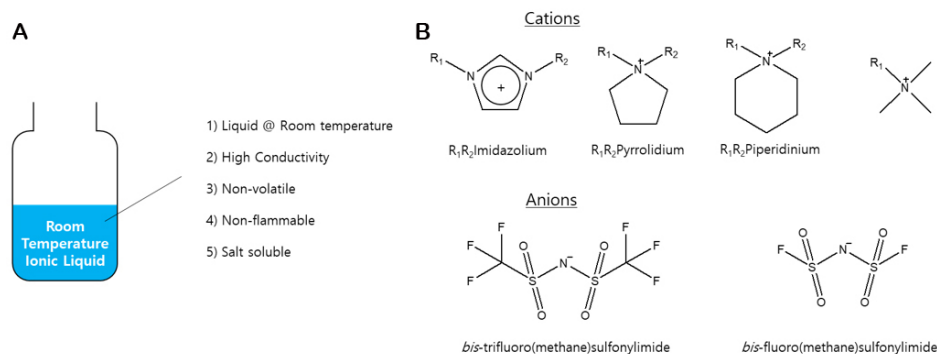


Figure 2. (A) Physical characteristics of ionic liquids. (B) Representative cation and anion structures for ionic liquids.

series are utilized^[24-27]. Depending on the binding affinity between the cations and anions, there are cases where they exist as liquids at room temperature and cases where they do not exist. The design of the cation and anion structures is aimed at reducing mutual Coulombic interactions by controlling the substituents. Electrolytes that dissolve lithium salts in glyme-based solvents are also categorized as ionic liquids because of their low volatility and low flammability^[28-30].

In LIBs, high-voltage cathode and low-voltage anode materials are used to achieve high energy densities. The general requirement for electrolytes in electrochemical systems is that they remain inert and do not participate in the electrochemical reactions. In some cases, such as in redox flow batteries, the electrolyte may participate in electrochemical reactions; however, this is an exceptional case. Therefore, for the stable operation of secondary batteries, the electrochemical stability of RTILs should encompass the operating voltage ranges of both the cathode and anode. The electrochemical potential window is determined by the HOMO/LUMO energy levels of the RTILs^[31-33]. Generally, the oxidative stability of ionic liquids is determined by the choice of anion, whereas the reductive stability is influenced by the cation. Recent studies have reported that the interplay between cations and anions can also affect electrochemical stability^[31].

However, even if the HOMO/LUMO levels of the electrolyte do not meet the voltage window of the battery, SEIs can stop unwanted reactions in the electrolyte. The performance of batteries can vary depending on the combination of the electrode and electrolyte; therefore, both the electrochemical stability and the electrode film formation mechanism need to be considered simultaneously. M. Ishikawa reported that when using an electrolyte based on 1-ethyl-3-methylimidazolium (EMIm⁺), in combination with lithium salts, in the presence of BF₄⁻ anions in TFSI⁻, FSI⁻, and BF₄⁻ combinations, the BF₄⁻ anions formed an unstable SEI on the lithium metal surface, while anions, such as FSI⁻ and TFSI⁻, were reported to form stable SEI on the lithium metal^[34,35]. The SEIs from FSI⁻ and TFSI⁻ effectively reduced the electrochemical reduction deposition of EMIm⁺ on RTILs. Thus, the SEI film on the negative electrode was highly influenced by the anionic species of the RTILs. Some chemical species, such as vinyl carbonates (VC), fluoroethylene carbonates (FEC), and vinyl ethylene carbonates (VEC), have been used to protect the anode by forming a stable SEI^[36-38]. These additives can also be used to stabilize RTILs on negative electrodes^[39-41].

For the cathode, the cation substituents were controlled to deliver a cathode-electrolyte interface (CEI). The long alkyl, allyl, and cyano groups were substituted with imidazolium, pyrrolidinium, and piperidinium to improve the CEI of the positive electrode and the electrochemical stability^[24,34,42,43]. Previous SEI analyses confirmed the presence of fluoride and organic films, indicating that both RTIL ions contributed to the formation of the SEI.

IONIC CONDUCTIVITY OF IONIC LIQUID

Ionic liquids are ionic materials that exhibit both ionic conductivity and solvent properties. However, lithium-ion conductivity is required for lithium secondary batteries, which distinguishes them from the conductivity of typical RTILs. Lithium salts, which contain the same anions as RTILs, are commonly used in combination with RTILs. In cases where different anions are used, they may not exist as liquids, depending on the type of RTIL used. Generally, RTILs containing imidazolium salts exhibit low viscosity and good ion conductivity^[31,44,45]. They can be liquids even with BF_4^- , which has smaller anions than TFSI⁻ and FSI⁻. However, imidazolium with FSI⁻ and TFSI⁻ exhibited lower viscosities and higher conductivities than those with BF_4^- . EMI⁺ with TFSI⁻ and FSI⁻ demonstrates the lowest level of viscosity and the highest level of conductivity (EMI-TFSI: $\eta = 34$, $\sigma = 14.4 \text{ mS cm}^{-2}$ & EMI-FSI: $\eta = 18$, $\sigma = 15.4 \text{ mS cm}^{-2}$, as shown in Table 1)^[46-51].

However, the use of imidazolium salts as RTIL electrolytes is limited because of their poor electrochemical stability. The presence of an acidic proton at the imidazolium 2-C position leads to poor electrochemical reduction stability. To overcome this problem, RTILs that incorporate cyclic or quaternary ammonium cations have been introduced. These compounds include pyrrolidinium, piperidinium, and ammonium. However, these structures lack conjugation within the cationic molecular framework, which results in inferior charge delocalization. Therefore, they only exist in a liquid state at room temperature when paired with bulky anions, such as TFSI and FSI, which have high charge delocalization. In addition, their conductivities and viscosities are inferior to those of imidazolium. Thus, to achieve asymmetry in the cation, one substituent attached to the nitrogen is often fixed as a methyl group, whereas the other is substituted with a relatively long substituent. However, it has been found that conductivity deteriorates when substituents larger than the propyl group are introduced. Among these alternative cations, pyrrolidinium is the most feasible cation candidate for RTILs because of its low viscosity and high conductivity (PMPyr-TFSI: $\eta = 59$, $\sigma = 4.9 \text{ mS cm}^{-2}$, as shown in Table 1).

STRATEGIES FOR MINIMIZING RESISTANCE OF BATTERIES

Sulfide-type solid electrolyte

Solid electrolytes used in SSBs can be classified as oxide- or sulfide-based. Between these two, sulfide-based solid electrolytes exhibit higher ionic conductivities than oxide-based solid electrolytes. Consequently, there has been a recent focus on research related to sulfide-based solid electrolytes^[52-54]. In 2015, Jung proposed the first SSB system using RTILs with anhydrous triglyme combined with Li-TFSI as an interfacial ion transport stabilizer. This system utilized a sulfide-based solid electrolyte, LGPS, and introduced the use of an ion-conductive liquid in SSBs^[55]. It has been observed that when tri-glyme is used in a molten state in the presence of lithium salts, it reduces the solubility and the reactivity of LGPS [Figure 3A]. This reduces the interfacial resistance between the LiFePO_4 (LFP) cathode particles and the LGPS solid electrolyte. In 2019, the same research group reported a slurry-based SSB electrode fabrication process using nitrile butadiene rubber (NBR), a polymer, and an RTIL to control the solvation environment and achieve high interfacial conductivity^[56]. Gao reported a stable cycle life of 2000 cycles at a current density of 2.5 C for a half-cell battery system utilizing LiG_3 -Li-TFSI RTILs with an LGPS electrolyte on a $\text{Li}_4\text{Ti}_5\text{O}_{12}$ anode^[57]. Fan improved the cyclability characteristics of $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ (NCM622) and Li metal batteries using a $\text{Li}_6\text{PS}_5\text{Cl}$ argyrodite electrolyte, which is also a representative sulfide solid electrolyte. They applied Li-TFSI/triethylene glycol dimethyl ether to the Li metal surface, forming a favorable LiF-based SEI and thus enhancing the surface stability of the Li metal electrode^[58]. In summary, many studies using glyme-based ionic liquids as electrolytes have been reported for sulfide-based solid electrolytes. In systems such as Li-S batteries, electrolytes, such as TEGDME, typically exhibit high interfacial resistance when not used with solid electrolytes, necessitating their combination with LiNO_3 to reduce polarization of Li metal electrodes.

Table 1. Viscosity and ionic conductivity with various structured RTILs

Cation	Substituent1	Substituent2	Anion	Viscosity (cP)	Conductivity (mS/cm)	Ref.
Imidazolium	Ethyl	Methyl	BF ₄ ⁻	27	14.4	[46]
	Propyl	Methyl	BF ₄ ⁻	58	7.2	[46]
	Butyl	Methyl	BF ₄ ⁻	76	4.3	[46]
	Isobutyl	Methyl	BF ₄ ⁻	76	1.8	[46]
	Allyl	Methyl	BF ₄ ⁻	45	10.0	[46]
	Ethyl	Methyl	TFSI ⁻	34	8.6	[49,50]
	Butyl	Methyl	TFSI ⁻	52	3.9	[50,51]
	Ethyl	Methyl	FSI ⁻	18	15.4	[47]
Pyrrolidinium	Propyl	Methyl	TFSI ⁻	59	4.9	[47]
	Allyl	Methyl	TFSI ⁻	52	5.7	[47]
	Allyl	Allyl	TFSI ⁻	57	4.6	[47]
	Propyl	Methyl	FSI ⁻	40	8.2	[47]
Piperidinium	Propyl	Methyl	TFSI ⁻	141	2.9	[47]
	Allyl	Methyl	TFSI ⁻	108	3.5	[47]
	Allyl	Allyl	TFSI ⁻	113	2.3	[47]
	Propyl	Methyl	FSI ⁻	95	3.7	[47]
Ammonium	isobutyl	Methyl	TFSI ⁻	100	0.017	[51]
	isobutyl	Methyl	FSI ⁻	53	5.8	[51]
	Allyl	Ethyl	TFSI ⁻	67	2.5	[48]
	Allyl	Propyl	TFSI ⁻	79	2.1	[48]

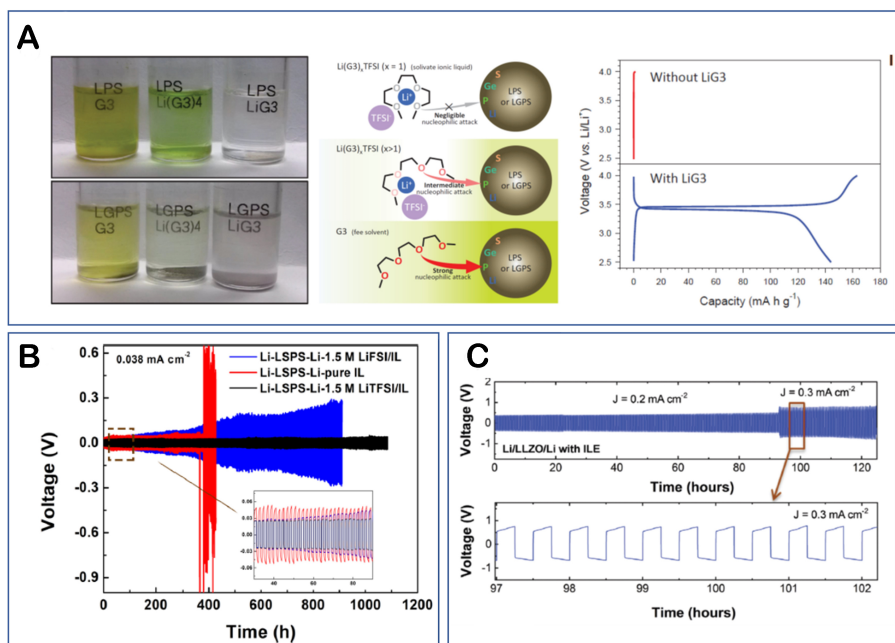


Figure 3. (A) Stability of tri-glyme ionic liquid electrolyte and voltage curves with/without ionic liquids^[55]. (B) Voltage curves of Li-Li symmetric cells with LSPS solid electrolyte and ionic liquids^[59]. (C) Cycle performance and Nyquist plot of RTIL-LiTFSI-PEO hybrid interphase material and NBR binder^[67].

However, in this SSB system, excellent performance was achieved even without LiNO₃, indicating that the optimal physical and chemical characteristics may differ from those of RTIL-based liquid phase

electrolytes^[29,30].

Yang reported the physical/chemical improvements of 1.5 M LiTFSI/N-propyl-N-methylpyrrolidinium (PMPyr) TFSI electrolyte on the lithium surface of the sulfide-based solid electrolyte $\text{Li}_{10}\text{SnP}_2\text{S}_{12}$. This study demonstrated a reduction in the lithium surface resistance, indicating improved performance in terms of physical and chemical properties [Figure 3B]^[59]. Cho utilized a solid electrolyte with minimal ionic liquid content on the surface of LGPS by separating the LGPS material from the N-butyl-N-methylpyrrolidinium (BMPyr) TFSI electrolyte after stirring. The study reported that LGPS exhibits high stability in pyrrolidinium-based electrolytes and reduces contact resistance, resulting in decreased solid electrode contact resistance in $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (NCM811) high-voltage, high-capacity cathode materials^[60]. A subsequent study reported a cyclability of 50 cycles, achieving performance comparable to that of high-pressure coin cells through the manufacturing method of NCM cathodes coated with polyethylene oxide (PEO) material and its composites in the form of slurry coating on LGPS solid-state electrolyte pellets [Figure 3C]^[61]. The combination of PEO and RTILs produces a less resistive interface in SSBs, based on Nyquist plots. By dissolving various types of salts, RTILs are readily accessible owing to their high level of safety. In line with this, recent research has also expanded to solid-state electrolytes using Na metal and Na_3PS_4 , Na_3SbS_4 , and Na-TFSI^[62-64].

Oxide-type solid electrolyte

There are many reports on the combination of oxide-type solid electrolytes and RTILs. Oxide-type solid electrolytes have more severe interfacial resistance problems owing to their lower conductivity than sulfide-type solid electrolytes. In addition, when pressure is applied to sulfide-based solid electrolytes, a relatively ductile interface is formed, resulting in a wide interface. However, it is difficult for solid oxides to form such wide interfaces, even under high pressure. In contrast, oxide-type solid electrolytes exhibit a wide electrochemical stability window^[65]. In this regard, RTILs are expected to mitigate the increase in resistance caused by subreactions at the oxide electrolyte interface. Owing to the rigid, solid nature of oxide electrolytes, the increased contact obtained from the lower interfacial resistance resulting from the formation of a liquid interface is expected to have a more significant effect. Traditionally, ceramic materials, such as Al_2O_3 or SiO_2 , have been used as supports for gel polymer composites that combine polymers such as PVDF and RTILs^[66-69]. It is known that the metal oxide surface aids in lithium-ion transport, leading to improved conductivity, and the combination of oxides with RTILs is a well-established research approach. In other words, the combination of metal oxides results in increased conductivity and enhanced mechanical strength, which has long been reported. However, with the increasing conductivity of oxide-type solid electrolytes, there has been a greater focus on the role of RTILs as enhancers of the physical electrode interface conductivity rather than solely improving the electrolyte conductivity.

Kohl reported the use of LISICON and 1.0 M Li-TFSI PMPyr-FSI as separator replacement materials for the stable operation of graphite/Li metal half-cell batteries even at high temperatures [Figure 4A]^[70]. Previously, it was reported that a PMPyr-FSI liquid electrolyte can form a stable SEI on both lithium metal and graphite electrodes in conventional LIBs^[71-73]. It has been found that the same type of FSI electrolyte can also form a stable SEI in such solid-type systems. Various high-conductivity oxide-based solid electrolytes, such as $\text{Li}_{0.33}\text{La}_{0.56}\text{TiO}_3$ (LLTO), $\text{Li}_{1+x}\text{Al}_x\text{Ti}_{2-x}(\text{PO}_4)_3$ (LATP), and $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO), have been discovered^[74-76]. As a result, a significant amount of research has been conducted on systems that combine high-conductivity solid electrolyte materials with RTILs to reduce interfacial resistance. Ogumi proposed an experiment involving direct contact between the electrode and the solid electrolyte by introducing an RTIL/LLTO interface. By controlling imidazolium and piperidinium cations and TFSI, BF_4^- , and BETI anions, it was reported that the anions have a more significant effect on the activation energy. Subsequently, RTILs were used to provide ion conductivity to the composite electrodes in SSB systems utilizing LLZO^[77]. Many solid-

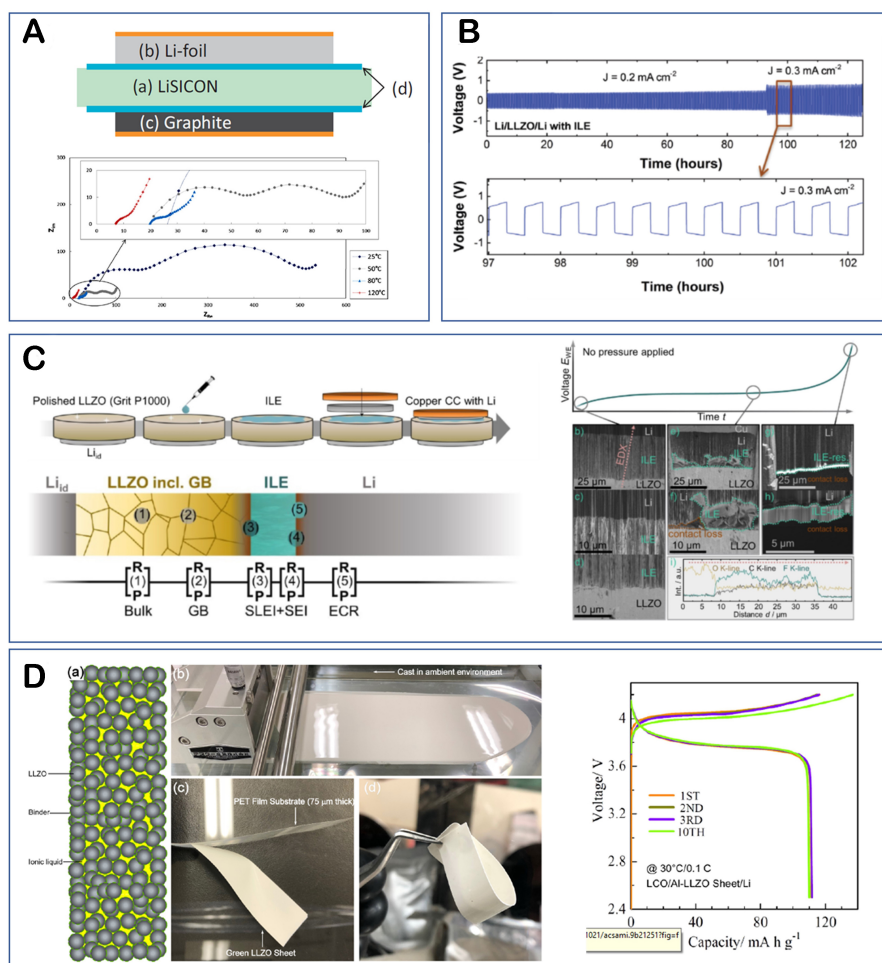


Figure 4. (A) Schematic figure of a LISICON separator half-cell with wetting agent^[70]. (B) Voltage curves of Li-Li symmetric cell with LLZO and ionic liquids^[79]. (C) Schematic figures of LLZO and ionic liquid electrolyte in SSBs and cross-sectional SEM images representing interfacial failure modes^[85]. (D) (Left) Schematic figure of the flexible composite Al-LLZO sheet and its fabrication procedure. (Right) Voltage curves from LiCoO₂ half-cells at 30 °C^[86].

state systems have reported the use of LFP as a cathode because RTILs are electrochemically inert at the high cutoff potential of the LFP cathode. Recently, Song reported a Li/LFP half-cell battery system utilizing 1-butyl-3-methylimidazolium bis (fluorosulfonyl) imide (BMIm-FSI) and LAMP^[78]. To mitigate the safety concerns associated with imidazolium-based electrolytes and Li metal, a multilayer concept was also employed in which BMIm-FSI was applied only to the cathode-side electrolyte. Passerini implemented a Li/LFP half-cell battery system using an electrolyte mixture of BMPyr-FSI and LiTFSI salt, which exhibited higher stability than imidazolium-based electrolytes. LLZO was used as the electrolyte to enhance the performance [Figure 4B]^[79]. RTILs with alkoxy-substituted FSI electrolytes in the piperidinium family have been reported to exhibit stable cycleability. By utilizing a 60 LAMP electrolyte, a relatively high-voltage NCM811 material, and a composite with BMPyr-TFSI, excellent cyclability results were achieved, maintaining a capacity retention of over 99.3% after 200 cycles^[80].

In addition to simply mixing RTILs and oxide electrolytes, research has also reported the enhancement of interfacial safety using LAMP oxide solid electrolytes in combination with PMPyr-TFSI and poly RTIL-gel^[81,82]. They also reported the use of BMPyr-TFSI, BMIm-TFSI, and EMIm-TFSI in conjunction with

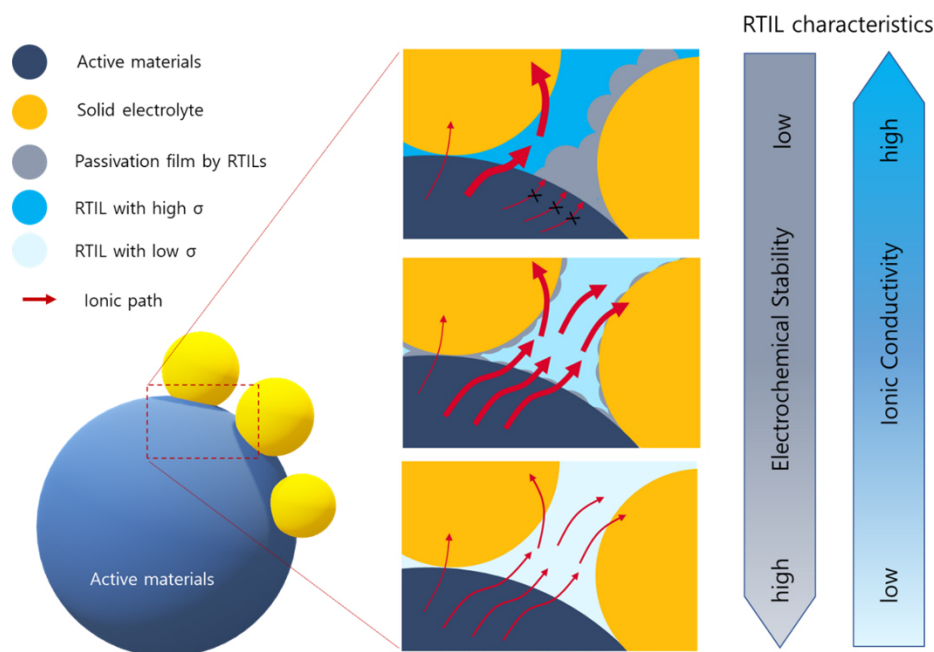


Figure 5. Schematic figure of interface among active materials/solid electrolyte/RTILs along electrochemical behaviors of RTILs.

PEO to enhance the interfacial safety of LLZO electrolytes^[83,84]. On the other hand, J. Janek reported the use of BMPyr-TFSI ionic liquid between LLZO and lithium interfaces, enabling Li-stripping of over 15 mAh·cm⁻² without additional cell pressure [Figure 4C]^[85]. Kanamura fabricated flexible electrolyte membranes by mixing LLZO with a binder using a slurry casting process with BMPyr-TFSI. They demonstrated that it electrochemically works in solid batteries by showing that the LiCoO₂ half-cells containing these composites had a specific capacity of 110 mAh g⁻¹ under 30 °C [Figure 4D]^[86]. After removing kinetic hindrances, their specific capacities were measured to be 140 mAh g⁻¹ at 60 °C. The highly resistive behavior at the solid interface of solid batteries remains an issue for the combination of RTILs and oxide-type solid electrolytes.

PERSPECTIVE & OUTLOOK

All-solid-state batteries, which do not have a conventional electrochemical liquid interface between the electrodes and electrolyte, generally suffer from high interfacial resistance. More critically, degradation at the electrode-SEI in all-solid-state batteries has been identified as a major factor that exacerbates the interfacial resistance. Under such circumstances, RTILs, which can form a liquid interface, have the potential to be applied in the manufacturing process of all-solid-state batteries without compromising safety owing to their non-volatility and electrochemical/chemical stability. Research on RTILs as ion-conductive media has been consistently conducted, building upon knowledge gained, particularly for LIBs with many combinations of active materials^[4,10,87-89]. Many RTIL/solid electrolyte composite results have been reported, and it is anticipated that interface stabilization through RTILs will continue to gain attention as they contribute to enhancing the completeness of all-solid-state batteries.

The development of novel RTILs that ensure interfacial stability with sulfide- and oxide-type solid electrolytes, along with the introduction of a new all-solid-state battery system, is expected to be a highly promising approach. Sulfide-based battery systems have been the subject of numerous studies owing to their high conductivity, which makes them suitable for all-solid-state battery systems. This is because the

high conductivity of the solid electrolyte enables more promising technological development towards all-solid-state batteries by reducing the contact resistance via highly conductive and safe RTILs contained within. However, developing new RTILs that are electrochemically and chemically stable in fragile sulfide-type solid electrolytes is necessary. New types of RTILs that lead to the stable passivation of electrode-active materials can be pursued. Additionally, the development of stable and conducting RTIL electrolytes capable of compensating for high interfacial resistance can serve as an approach to reducing solid/solid interfacial resistance [Figure 5].

In contrast, oxide-based systems have relatively low conductivities but exhibit high chemical stability. Thus, the application of RTILs in oxide systems is expected to be relatively feasible. Owing to the low electrolyte conductivity of oxide-type solid electrolytes, the use of RTILs is anticipated to be more prevalent to compensate for poor ion conductivity. In addition to reducing the interfacial contact resistance, it is expected that research on various electrode chemical interfaces will lead to the development of more complete oxide-RTIL electrolyte systems in the future. Additionally, because the liquid interface is introduced as an additional ion transport medium in all-solid-state batteries, it is expected that issues related to the viscosity and conductivity of RTILs will receive less attention than stability concerns. Once the polarization issues at the solid interface are addressed through novel RTILs, further attention must be paid to addressing the subsequent minor issues that are intricately involved in battery operation. These secondary issues include the corrosiveness, wide operating temperature range, and flammability of the RTILs. In addition to the RTILs used in conventional LIBs, new RTIL systems suitable for all-solid-state batteries are being actively investigated.

DECLARATIONS

Authors' contributions

Proposed the topic of this review: Mun J

Prepared the manuscript: Jeong S, Ho VC, Kwon O, Park Y, Mun J

Collectively discussed and revised the manuscript: Jeong S, Ho VC, Kwon O, Park Y, Mun J

Availability of data and materials

Not applicable.

Financial support and sponsorship

This research was financially supported by the Institute of Civil Military Technology Cooperation, funded by the Defense Acquisition Program Administration, and the Ministry of Trade, Industry and Energy of the Korean government under grant No. UM22213RD2.

Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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