



Review article

Different roles of ionic liquids in lithium batteries

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HIGHLIGHTS

- All possible applications of ionic liquids in lithium batteries are reviewed.
- Potentials and limits of ILs for LIBs are described.
- Superior possibility of ionic liquid crystals as future electrolytes is presented.

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ABSTRACT

Ionic liquids are often named solvents of the future because of flexibility in design. This statement has given credence that ionic liquids should simply replace the problematic electrolytes of lithium batteries. As a result, the promising potentials of ionic liquids in electrochemical systems are somehow obscured by inappropriate expectations. We summarize recent advancements in this field, especially, ionic liquids as standalone electrolytes, additives, plasticizers in gel polymer electrolytes, and binders; and attempt to shed light on the future pathway of this area of research. Ionic liquids are not dilute media to serve as pure solvents in electrochemical systems where mobility of ions is the priority; instead, they can contribute to the ionic conductivity of various components in a battery system. Owing to the enormous possibilities of ionic liquids, it is not merely a matter of choice. Ionic liquids can be used to design novel types of electrolytes for a new generation of lithium batteries. A promising possibility, which is still at a very early stage, is *supercooled ionic liquid crystals* for fast ion diffusion through the guided channels of a liquid-like medium. This, of course, will be a breakthrough in the realm of electrochemistry, far beyond lithium battery field, when materialized.

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1. Introduction

The pioneering works of Humphry Davy for electrolysis of molten salts paved the path for electrochemistry in molten electrolytes [1], but the focus was practical for the preparation of reactive metals in elemental form. The next step was taken by Charles Martin Hall for the preparation of metallic aluminum *via* electrolysis of aluminum oxide dissolved in cryolite [2]. Due to the industrial importance of this method (which is still the primary approach for the production of aluminum), this idea was widely followed for industrial extraction of metals. The key feature of this idea was the possibility of forming eutectic molten salts to conduct

an electrolysis at lower temperatures.

Molten salts were subject to numerous studies from fundamental electrochemistry to various applications (metallurgy, fuel cells, *etc.*). Nevertheless, molten salts are usually the electroactive materials in electrochemical systems, exactly as it was in Davy's works. Ionic liquids (ILs) are indeed a game changer in electrochemistry by bringing new opportunities [3–9]. This is the main difference of molten salts and ILs; the former is the main point of interest in an electrochemical system, but the latter provides a new set of media for electrochemical reactions. Thus, ILs can find a universal place in the realm of electrochemistry.

From the chemistry point of view, ILs are ionic salts with large cations (usually organic) and anions (commonly both organic and inorganic), which make the ionic forces weaker, resulting in lower melting points. By definition (just common agreement indeed), ILs are molten salts with melting point less than 100 °C [10], and sometimes ILs with melting points lower than 25 °C are exclusively

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named room temperature ionic liquids (RTILs). This definition is subject to controversy due to the artificial threshold which does not have scientific significance. In the absence of any standard definition, the present one is commonly accepted, though, there are some misleading in the terminology of ILs.

The interface of molten salts and ionic liquids is chloroaluminate, *i.e.*, something in-between. Aluminum chloride has weak ionic bonds, and thus, a relatively low melting point (192 °C, if avoiding sublimation under pressure), but it has no noticeable ionic conductivity, as ions are not free to move in the corresponding molten medium. Eutectic mixtures of aluminum chloride with alkali metal chlorides have melting points as low as 70 °C [11,12]. The concept of eutectic molten salts is simple: introducing more ions to the system breaks the ordered structure of ionic solid resulting in a lower melting point. This also provides more freedom for ions to transport within the molten medium, and thus, improves the ionic conductivity [11,12]. In the best case, chloroaluminates are something between ionic and molecular liquids, and thus, they have been called quasi-molten salt [13]. Chloroaluminates have also been utilized as the first room temperature molten electrolytes of alkali metal batteries too [13–15], but they are not practical choices for commercialization. Another critical issue is instability of chloroaluminates in the presence of moisture.

In any case, the attractiveness of low-temperature molten salts paved the path for the development of ILs in which the ionic bonds are weak, and thus, the melting point is low; but not at the cost of losing ionic characteristics, contrary to chloroaluminates. It was common to emphasize non-chloroaluminates ILs in the literature, but in today's terminology, ILs are simply referred to the latter class. In other words, when claiming the unique roles of ILs in future technology, this is about organic ILs, not chloroaluminate ILs.

It should be emphasized that ILs have distinctive differences with molten salts, and it is not appropriate to categorize ILs as a subgroup of molten salts by simply setting the melting point.

- (i) Almost all ILs are capable of forming supercooled liquids below their melting points. This is indeed a common mistake in the literature; the liquidity of ILs should not be just judged by their melting points but the glass transition, as many ILs with melting point above room temperature are liquid at room temperature [16].
- (ii) Almost all ILs are subject to ionic pairing and subsequent self-aggregation due to large asymmetrical ions. In fact, the essential characteristic of ILs is not merely weak ionic bonds, but unusual charge distribution over ions, making the system much more complicated than molten ionic salts. This feature can be better understood in the transition from ionic liquids to (ionic) liquid crystals, in which long ions are indeed agglomerated in oriented directions.

The growing interest in ionic liquids is enormous, and various aspects of ionic liquids have been extensively reviewed [9,17–33].

For lithium batteries, which are the topic under consideration in this review, a common strategy is to test available ILs simply to replace problematic organic carbonates. It is impossible to find the best choices out of 10^{18} possible ILs [10] empirically. Nevertheless, only a few ionic liquids have been used for various applications in lithium batteries (Table 1). On the other hand, it is not wise to focus on adapting ILs to do the job of available organic carbonates, because of primary advantages such as inflammability. ILs have lots of additional capabilities to do better jobs. Apparently, this possibility comes at a price: ILs are too complicated (in comparison with

Table 1
Physical properties of common ILs for electrochemical systems. References in the last three columns point to representative papers (reference number) used the corresponding ILs for lithium battery applications as pure electrolytes, mixed with additives, or in gel polymer electrolytes, respectively.

Cation	Anion	T _m (K)	T _g (K)	Window (V)	σ _c (S m ⁻¹)	η (cp)	Pure	Mix	Gel
EMI	Cl	362.15 [377]		5 [378]	0.343–3.709 [379]		[380]		
	BF ₄	288.15 [379]	178 [381]	4.3 [382]	1.38 [383]	37.7 [382]	[59,80,204]		[232,383,383,384]
	TFSI	258.15 [385]	175.15 [385]	4.3 [386]	0.86 [387]	34 [388]	[74,76,93,104,150,151,387,389–394]	[157]	[395–398]
	BETI	272.15 [386]	188 [386]	4.1 [386]	0.34 [387]	61 [386]			[399]
	MSI	223.15 [385]		2.5 [385]	0.017 [389]	787 [385]			
	OTf	264 [388]		4.1 [388]	1.1 [387]				[400]
	TA	259 [388]		3.4 [388]	0.96 [390]	35 [388]			
	F(HF) _{2,3}			3.3 [401]	12 [402]	4.9 [403]			
	FSI						[63,78,126,402,404–407]		
	FSA						[73]		
	TFSA						[408]		
	PF ₆							[409]	
	DCA	252.15 [410]	169.15 [410]						
BMI	BF ₄	190.65 [412]	188.15 [413]	6.1 [414]		219 [415]	[416]		[417]
	TFSI	269.15 [388]	169.15 [415]	4.76 [417]	0.39 [390]	52 [388]			[417]
	PF ₆	283.15 [415]	193.15 [415]	5 [414]	0.1 [418,419]	450 [415]			
	TA	233.15 [388]		5.7 [414]	0.32 [390]	73 [388]			
C8MI	TFSI		189.15 [420]	4.83 [417]		119.3 [417]			
C10MI	TFSI	244.15 [420]	190.15 [420]	4.89 [417]		152.8 [417]			
MDI	BF ₄			3.8 [421]					
	TA			5.7 [414]					
	TFSI			4.76 [417]					
	PF ₆			6.35 [417]					
M1,2E3I	TFSI			4.4 [388]		88 [388]			
	DMPi	TFSI		5.2 [418]	0.3 [389]	60 [385]			
P13	Me			5.37 [422]	0.046 [423,424]				
	MSI	210.15 [425]		4.25 [425]					
P14	FSI						[63,78]		
	TFSI	285.15 [426]	183.15 [426]				[70,170,394,424,427,428]	[203,224,429]	[253,398]
	PF ₆							[409]	
	FSA						[73]		
P14	TFSI	255.15 [425]	186.15 [425]	6 [430]	0.22 [375]	85 [430]			
	MSI			4 [425]		1680 [425]			

common solvents), and we are far from a satisfactory understanding of IL-based systems. This review aims to shed light on the roadmap for utilizing ILs for the next generations of lithium batteries.

2. Electrochemistry in ionic liquids

Owing to the ionic conductivity of ILs, they can be directly used as electrolytes in various electrochemical systems [34–37]. The interesting feature is that ILs can be used as a one-component electrolyte (of course, not applicable to lithium batteries, as no practical IL with Li cation is known), whereas electrolytes in most electrochemical systems are three-component: solvent, supporting electrolyte, electroactive species. However, this is not for simplicity, as electrochemistry in ionic liquids is much more complicated and different from conventional electrochemistry.

It can be stated that almost all equations derived in electrochemistry textbooks are invalid in ILs. Electrochemical equations are usually obtained by solving basic equations (such as Fick's law, Navier-Stokes, etc. [38–40]) of fluid mechanics under simplified conditions. However, the assumptions made in conventional electrochemistry are no longer valid in ILs. In fact, diffusion in ILs is not as straightforward as that in conventional electrolytes, as it is more like solid-state diffusion in which conventional electrochemistry is not valid [41–43]. One may consider diffusion in ILs as something in-between of conventional electrolytes and solid state, but it can be even more complicated than solid-state diffusion due to the ever-changing pathways of diffusion as a result of frequent ion pairings [44–49]. We do not aim to discuss the electrochemistry of ILs deeply here; just highlighting the major differences, which should be taken into account. Diffusion of electroactive species is usually the most important process in an electrochemical system such as lithium batteries. To clarify possible issues, let us review the validity of common assumptions made in conventional electrochemistry for ILs.

- (i) *Diffusion is one-dimensional towards the electrode surface* (as a result of concentration gradient formed by redox reaction at the electrode surface, consuming reactants). One-dimensional diffusion can be assumed in a fluid in which the electroactive species can find the shortest path (straight line) toward the electrode surface (Fig. 1a). This is not possible in ILs at least for two reasons: (a) ILs are usually viscous fluids, and thus the straight path for the diffusion of electroactive species is not simply provided. (b) ILs are wholly composed of large ions. Therefore, there are blocking interactions between the electroactive ions and IL ions (Fig. 1b). Electroactive species colliding IL ions can be bounced back or significantly deviated from straight direction. While conventional solvents are somehow inert media (electrostatically solvating the charged ions), the IL medium is strongly interactive. On the other hand, as illustrated, the shape of IL ions can be unconventional and asymmetrical, which can affect the interactive pathway of diffusion too. In a sense, the shape dependency of diffusion pathway in ILs is somehow similar to the influence of the geometrical structure of porosity on solid-state diffusion channel. With a similar concept, porous liquids are made of ILs [50].
- (ii) *Diffusion is non-interactive.* In an electrochemical system, reactants are consumed at the electrode surface resulting in the formation of a concentration gradient. Diffusion of electroactive species from bulk electrolyte towards the electrode surface is driven by this concentration gradient. To apply basic diffusion equations, this diffusion should be a physical process to satisfy the concentration gradient. This

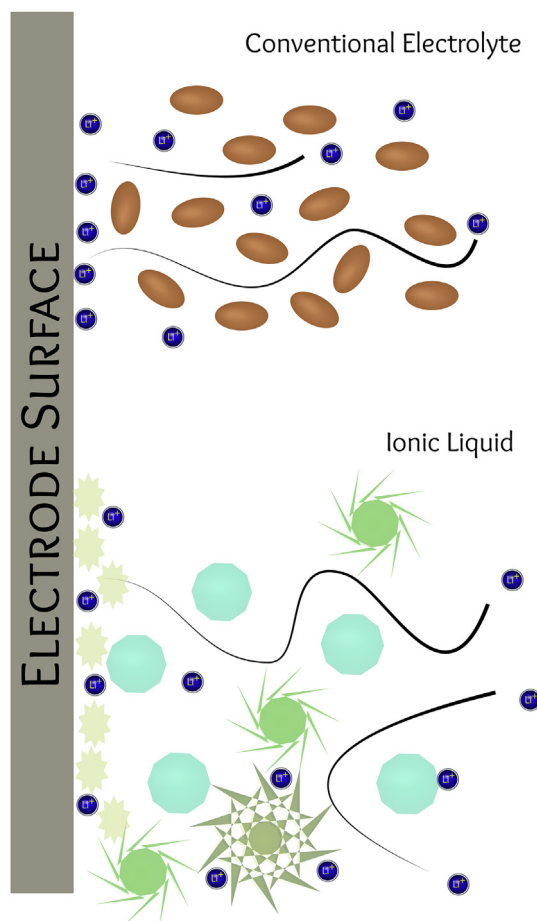


Fig. 1. Schematic illustration of (a) one-dimensional diffusion in conventional electrolytes, and (b) complicated interactions of diffusion in ILs. The branches of IL ions show a possible asymmetrical arrangement of interactive groups. Furthermore, IL ions can be chemically/physically sorbed on the electrode surface as well as Li ions.

assumption is possible for conventional electrolytes, as the electroactive species can diffuse within the inert medium without major interactions. However, in ILs, electroactive species can be subject to active chemical interactions with the IL ions in the way of reaching the electrode surface (Fig. 1b). Therefore, the transport of electroactive species is not solely governed by the gradient-driven diffusion.

- (iii) *To dominate the role of diffusion in the mass transport equations, the possibility of migration should be neglected.* In conventional electrochemistry, this is achieved by using excess supporting electrolyte. Thus, the ratio of the electroactive ions to the system ions is negligible. In an IL, the entire system is electroactive species.
- (iv) *To neglect the role of the natural convection, diffusion should be fast enough to conduct the experiment in a relatively short time* (e.g., under 100s in a chronoamperometric experiment). This could be assumed in conventional electrolytes where the diffusion coefficient is in order of $10^{-5} \text{ cm}^2 \text{ s}^{-1}$, but not in ILs where the diffusion coefficient is in order of $10^{-9} \text{ cm}^2 \text{ s}^{-1}$ (at least for practical cases like lithium batteries in which concentrated electroactive species are used).
- (v) *Complication of a changing structure.* Although it is still subject to preliminary studies [44], it is believed that ion pairing/de-pairing of ILs is very complicated resulting in frequent changes in the IL structures. In conventional

electrochemistry, solvated ions can be considered as permanent structure; but in ILs, solvating ions are in interaction with temporary agglomerated ion bundles.

The first case makes diffusion in IL more like solid-state diffusion, but the other cases make it even more complicated. In general, the purpose was to clarify the limitation of available equations to explain electrochemistry in ILs. In other words, we need profound fundamental studies to understand fully electrochemical systems utilizing ILs.

Moreover, as hypothetically illustrated in Fig. 1, IL ions can chemically interact with the electrode surface as well as Li ions. This can block the surface porosity where Li solid-state diffusion starts.

3. Pure ionic liquids as electrolytes

Since the commercialization of lithium batteries, the dominant electrolytes were based on organic carbonates. Despite their considerable advantages, they have severe problems, particularly from the safety perspective. In the past three decades, numerous efforts have been paid to this issue to find potential alternatives to available non-aqueous solutions. On the other hand, 5-V cathode materials have been extensively developed [51–62], but limited by the lack of appropriate electrolytes having electrochemical stability at high anodic potentials, and thus, there is an active quest for finding appropriate electrolytes for 5 V lithium batteries [56–61]. A possible solution is solid electrolytes [60,61], but the ionic mobility is a serious issue. Potentially promising candidates are ILs [62].

The unique features of ILs are frequently repeated in the literature as non-flammability, non-toxicity, low vapor pressure. Thus, many researchers attempted to replace current electrolytes with available ILs [62–92]. Interestingly, some electrode materials, which are not working in conventional electrolytes, showed promising performance in ILs [71,72,92]. Despite impressive results reported in the literature, there is still no practical potential to use ILs as standalone electrolytes in lithium batteries. In the absence of a comprehensive knowledge of the electrochemical processes involved in IL-based lithium cells, it is difficult to find the most appropriate ionic liquid for a particular system.

There are four key issues for ILs as sole electrolytes of lithium batteries, which are not necessarily disadvantageous but should be taken into account before developing any IL-based lithium battery.

High viscosity and slow diffusion: Viscosity of ILs is a great deal in electrochemical systems including lithium batteries [73,93–99], as it is connected with conductivity, diffusivity, and wettability; but this is more complicated than conventional fluid viscosity. This is of double importance, as adding lithium salt to an IL increases its viscosity and reduces conductivity. As a matter of fact, diffusion in ILs is not as slow as assumed. The diffusion coefficient of electroactive species in ILs can be in order of 10^{-7} cm² s⁻¹ showing diffusion-controlled behavior [100]. The problem of slow Li diffusion in IL is due to high concentration of lithium salt used in lithium batteries to increase the transference number. In other words, the viscosity is not the primary factor in slowing down the diffusion, but the active interaction of Li ions in the rigid structure of ILs.

Normally, diffusion of Li ions within electrolyte is much faster (even in viscous ILs) than solid-state diffusion within the electrode (anode or cathode). Thus, Li ions are always ready for the insertion process at the electrode surface. A critical issue of high viscosity is weak wettability of electrode materials in ILs [84], as the electrolyte should be spread within the porous structure of the electrode material. One may think of optimized porosity sizes of the electroactive material to improve wettability in a specific IL.

This failure has indeed overshadowed the superiority of ILs as

promising electrolytes. When the solid-state diffusion is much slower than Li diffusion through IL, like the case of LiFePO₄, the battery performance in IL is much better than that in conventional organic solutions [65]. In the case of fast solid-state diffusion within cathode materials such as LiMn₂O₄, the slow diffusion in ILs is the rate-determining step. For a similar reason, the failure of ILs for fast-charging cathode materials (such as LiMn₂O₄, LiCoO₂, etc.) appears at high rates, and when the Li ions have enough time to reach the solid electroactive material under slow charging/discharging, ILs fairly compete with conventional organic solutions [65].

It is worth considering that although low viscosity is in favor of rate capability, ILs with higher viscosities may deliver better battery performance in term of specific capacity [78].

Solvation in IL: The key step in Li insertion is the release of Li ions at the electrode/electrolyte interface [92]. Li solvation within an ionic liquid is significantly different from that by polar molecules [69,101–122]. At the electrode/electrolyte interface, lone Li-ions should be released from the solvation trap. Depending on the IL anions, breaking the solvation trap can be easier for Li ions, leading to a better battery performance [71]. However, it is impossible to make a general comparison between ILs and organic solutions, as the solvation can have an entirely different structure in ILs. It also should be taken into account, when referring to Li diffusion, it is not free diffusion of individual Li cations, but solvated Li ions. This means that diffusion process strongly depends on the size and charge of solvated Li, which are significantly dependent on the IL utilized.

In this case, lithium coordination in a specific IL plays a substantial role in the battery performance. Coordinations of lithium in protic and aprotic ILs are significantly different [122]. In general, lithium coordination number is lower in protic ILs. This adds an advantage for protic ILs, as it has been claimed that protic ILs can deliver all natural benefits of aprotic ILs for lithium batteries [123]. In general, aprotic ILs provide more stable and wider electrochemical window [9].

Double layer barrier at the electrode surface: In any electrochemical system, the electrode surface is capable of forming an electrical double layer by the charged species; but this phenomenon is more severe and complicated in ILs which are entirely composed of charged ions. This causes a critical inhibition for the insertion of Li cations to transfer through this barrier at the electrode surface (as illustrated in Fig. 1). Surface analyses have shown that the ILs do not form classical double layers; instead, complex interfacial layers composed of several layers of cations and anions are formed at the electrode/electrolyte interface [44,124–126].

Solid electrolyte interphase (SEI): Not only for IL-based but also for all lithium batteries, the formation of SEIs on the electrode surfaces (both anode and cathode) is a critical issue [127–143]. This solid layer can be helpful or harmful. Formation of a thin permeable membrane protects the electrode during the battery performance, but this layer can be blocking too. In organic solutions, SEI is typically formed by the decomposition of the organic compounds at extreme potentials. However, SEI in ILs is indeed the electrode passivation as a result of a chemical reaction [70,144–157], because the electrode surface can also be passivated electrolessly. This provides more flexibility for ILs to form protective SEI. In this case, the initial capacity in IL is usually lower, but the resulting SEI can provide an excellent cyclability [145].

Electrochemical reduction of IL cation can result in the formation of SEI, but this cannot be solely related to the electrochemical stability of the cation. In the case of EMI, which is a promising cation of ILs for lithium batteries, the corresponding anion plays a substantial role. Fig. 2a depicts while the system delivers the capacity and cyclability comparable with conventional organic

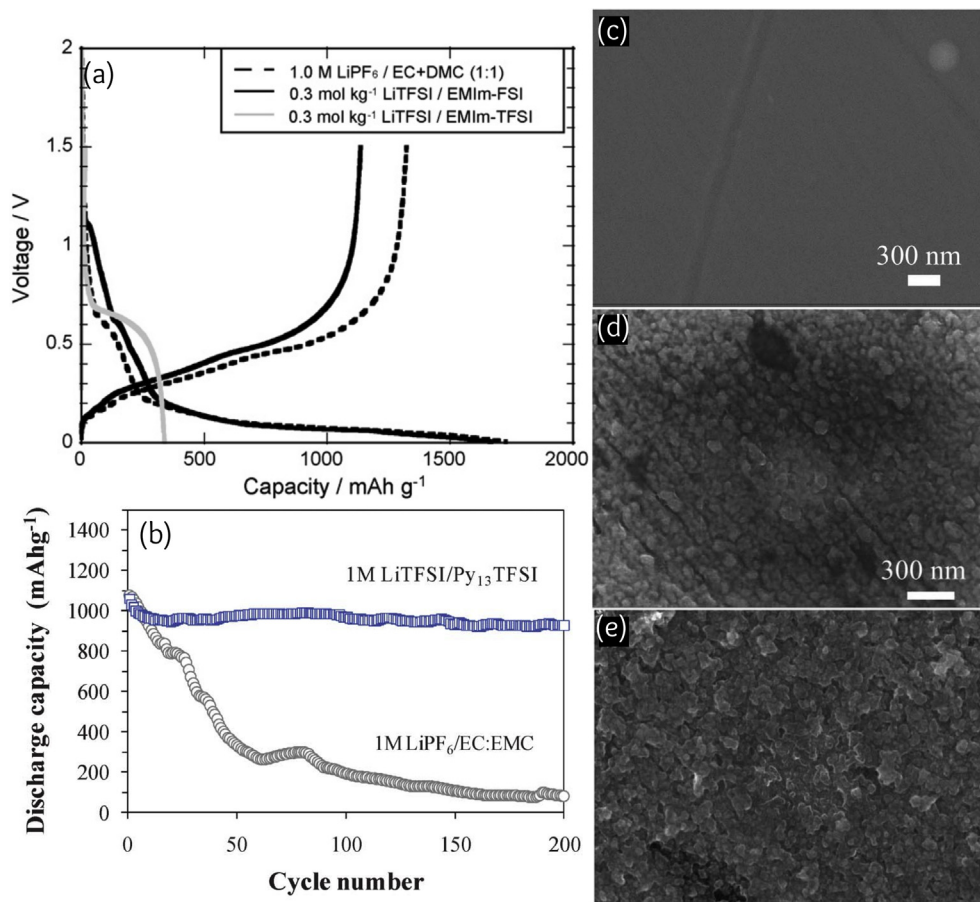


Fig. 2. (a) Charge–discharge profiles of Si–Ni–carbon composite electrode at the first cycle with 0.3 mol kg^{-1} LiTFSI/EMI-FSI, 0.3 mol/kg LiTFSI/EMI-TFSI, and 1.0 M LiPF₆/EC DMC at charge–discharge current of 500 mA g^{-1} . Reprinted from Ref. [68]. (b) Cyclability plot and SEM images for the SiO_{1.3} electrodes subjected to cycling in 1 M LiTFSI/Py₁₃TFSI and 1 M LiPF₆/EC:EMC, and (c,d) SEM images of the electrode cycled in the same electrolytes, respectively. Reprinted from Ref. [70].

solutions (*i.e.*, LiPF₆ in EC/DMC) when the anion is FSI, but negligible capacity can be achieved with TFSI [63,68,146].

In fact, anions of ILs are of vital importance in lithium battery application, though, cations are known as the main characteristic of ILs (note that ILs were preliminarily developed by increasing the size of cations). Although cathodic instability of ILs at negative potentials is due to cation reduction, it is the anion which plays a substantial role in the electrochemical stability and consequently wideness of the potential window (Fig. 3).

This indeed adds to the complicated nature of ILs, as the battery performance is generally affected by the cation rather than anion [158]. The reason (at least in ordinary cases) is that cations have more significant influence on the viscosity of Li-IL binary electrolytes.

It should be emphasized that the formation of a SEI is the result of the electrochemical reaction of both anions and cations at the electrode surface. Thus, the nature of a SEI is directly connected with the interfacial charge transfer at the electrode surface resulting in the film growth. There is no generalization that ILs are not capable of forming a stable and electrochemically active SEI, as this strongly varies from case to case, depending on the electrode material under consideration. As a result, some electrode materials show better cyclability in ILs rather than conventional electrolytes, as SEIs could not be formed at those specific electrode surfaces in the latter medium (Fig. 2b).

Another interesting feature of IL electrolyte can be seen by morphological changes caused by cycling. Obviously, the electroactive film is subject to roughening as a result of solid-state

diffusion in the course of charge/discharge (Fig. 2c–e). While the surface morphology is subject to severe changes after cycling in a conventional electrolyte, the electroactive film retains its dense structure after cycling in an IL electrolyte (Fig. 2d). The porosity formed after cycling in a conventional electrolyte results in lower mechanical stability (Fig. 2e).

Electrochemical reaction at the electrode surface is not only controlled by the electrode materials but also its atomic structure. In battery performance of similar carbon anodes, those with more edging carbon atoms display higher irreversible capacity due to side reactions [126]. For the case of dangling carbon atoms at the edge of graphene plane, it has been demonstrated that they change the charge transfer pathways in electrochemical systems [159].

According to the experimental results reported in the literature, it is unlikely to develop conventional lithium batteries with pure IL electrolyte. However, in this case, the first criterion is to find a low viscous IL [63], as apparently, the blocking SEI formed in high viscous ILs is much more stable resulting in irreversible capacity fading. In other words, the main problem of viscous IL is not slow diffusion in this case.

Besides the high-cost issue, pure IL electrolyte can be a practical potential for high-temperature batteries in which the viscosity is not significant. An R&D example is the development of sodium ion battery based on pure IL electrolyte by Sumitomo Electric Industries for high-temperature performance [160]. On the other hand, a characteristic feature of IL is unbeatable thermal stability in comparison with molecular solvents.

In addition to practical applications, pure electrolytes are of

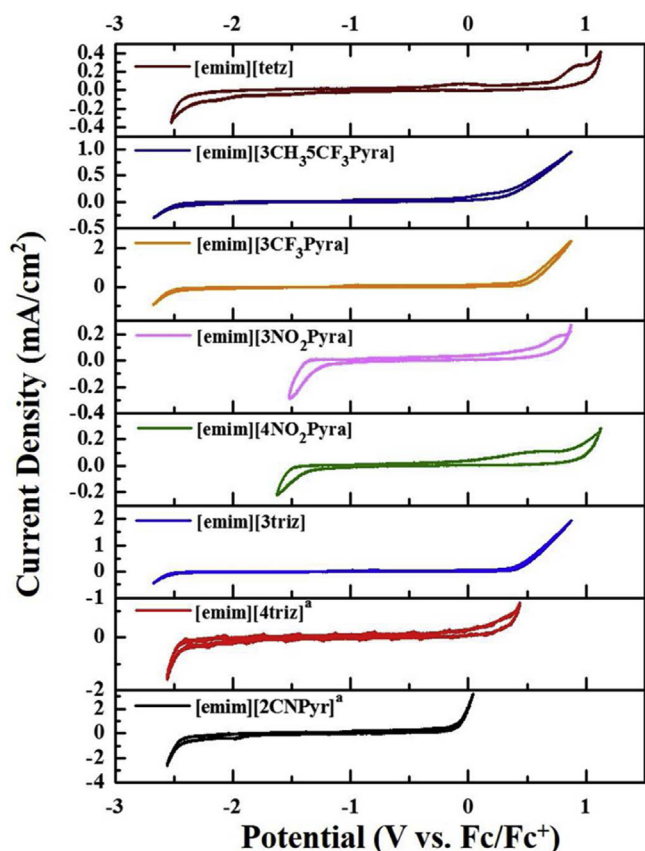


Fig. 3. Electrochemical windows of EMI-AHA ILs on glassy carbon at room temperature. Scan rate 100 mV s^{-1} . Reprinted from Ref. [96].

interest for fundamental studies of electrode materials to improve their battery performances regardless of their interactions with the electrolytes. For instance, aqueous solutions, which were of no practical interest, have been widely used to investigate electrochemical behavior of electroactive materials for lithium batteries [161–169]; now there is an emerging interest for practical applications of aqueous electrolytes due to low cost. Albeit, vague visions about electrochemistry in ILs have already restricted this possibility.

4. Mixture of ionic liquids and conventional electrolytes

As discussed above, current ILs are not appropriate candidates for standalone electrolytes of lithium batteries, but this is indeed an unjustified expectation. Each component of lithium batteries is improved by the aid of some additives. Hence, ILs also need some additives to overcome natural disadvantages. The simplest choices are organic carbonates, which are currently the dominant electrolytes of lithium batteries. It can also be seen this way too, that ILs are used as additives to improve the performance of available electrolytes [170–173].

Mixing ILs with organic carbonates can gain some advantages of both parties [76,174–201]. This is not the ultimate goal of replacing current organic solutions, as even their co-presence can make the cell vulnerable to safety issues. However, if the content of IL is high enough in this mixture, the electrolyte is practically non-flammable, as IL acts as a flame extinguisher [200]. As can be seen in Fig. 4b, adding 50% IL to a conventional electrolyte can significantly reduce the organic electrolyte flammability while the battery performance remains intact (Fig. 4c).

The specific issue of ILs is high viscosity which is accompanied by a relatively low ionic conductivity too. Mixing ILs and organic carbonates can lead to an optimum viscosity and conductivity. Interestingly, the ionic conductivity of the resulting mixture can be even higher than that of the organic carbonates [182]. Besides these obvious physical improvements (viscosity and conductivity), adding organic carbonates to ILs can make the following contributions to a better battery performance:

- (i) Although the nature of SEI is very complicated and not yet understood even in conventional electrolytes, it is believed that the SEIs formed in pure ILs are not electrochemically permeable for the diffusion of ions. Electrochemical impedance spectra show that the interfacial resistance of both anode and cathode decreases by adding an organic carbonate to the IL electrolyte (Fig. 4a).
- (ii) Along with a less-resistive SEI, organic carbonates significantly enhance the wettability of electrodes in the corresponding electrolytes. This directly improves the battery performance to achieve higher capacities in comparison with pure IL electrolyte (Fig. 4c).
- (iii) In general, ILs provide better battery performance for cathode rather than anodes, and still the main issue is on the anode side. Although ILs have wide electrochemical windows, they suffer from cathodic instability at negative potentials close to the standard potential of Li. As a result, ILs cannot be used with Li metal anode and other low-potential anodes. On the contrary, organic carbonates decompose at anodic potentials (this is the reason that they are not suitable for 5 V batteries) but have excellent stability at negative potentials. The presence of organic carbonates can somewhat extend the electrochemical stability of ILs towards negative potentials (Fig. 4d). Ideally, the system should remain stable with no decomposition process until reaching the standard potential of lithium in which Li electroplating occurs [178]. The electroplating process in pure IL electrolyte is usually weak due to high viscosity and low conductivity, and this can be used as a signal for finding an appropriate IL in which electroplating can be successfully conducted, and this is the reason that ILs are promising electrolytes for electrodeposition of metals, which cannot be electrodeposited from aqueous or organic solutions [31,202].

Mixing the electrolyte does not necessarily need comparable volumes of both components, as one can be iteratively an additive. A small amount of organic carbonates (e.g., 5%) can significantly improve the battery performance of ILs [203,204]. Adding 10–20% of organic carbonates can overcome the common problem of ILs including viscosity and ionic conductivity [186], while the electrolyte is still non-flammable. Molecular dynamic simulations have shown that the presence of organic additives increases the ionic mobility by lowering the lithium coordination [112].

In addition to adding organic carbonates to improve the battery performance of ILs, it is a practical approach to add ILs to available electrolytes of lithium batteries. In these cases, ILs are not treated as liquid electrolytes but organic ionic salts to introduce large ions into the electrolyte matrix. IL flame-retarding additives can significantly reduce the flammability of conventional organic carbonate electrolytes [205]. A small amount of ILs can improve the electrochemical stability of organic carbonate electrolytes for 5 V performance [172].

5. Gel polymer electrolytes

Besides the points mentioned earlier, there is always a severe

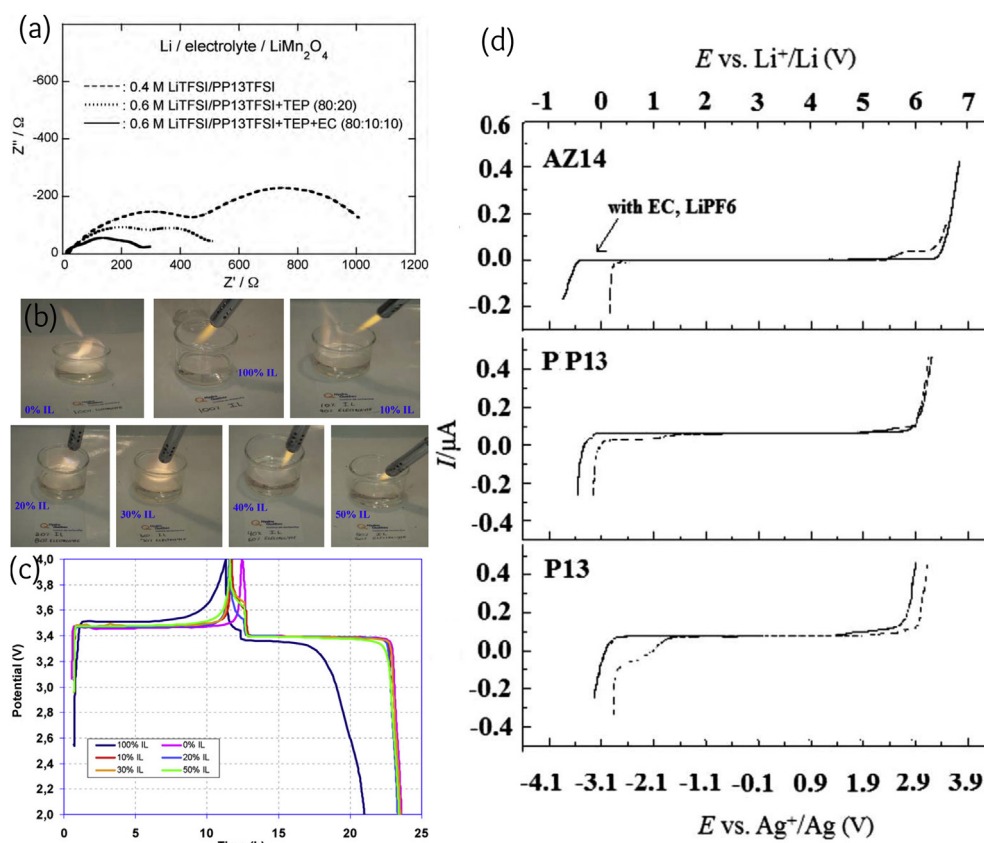


Fig. 4. (a) Electrochemical impedance studies of Li/LiMn₂O₄ cells utilizing different electrolytes. Reprinted from Ref. [186]. (b) The influence of IL (EMI-TFSI) on the flammability of a mixture of carbonates (EC/DEC/VC), and (c) charge/discharge behavior of a LiFePO₄ electrode in 1M LiPF₆. Reprinted from Ref. [181]. (d) Linear scan voltammetry of neat ionic liquids and their LiPF₆/EC (1M LiPF₆/50% EC) electrolyte using Pt working electrode and Ag wire reference electrode. The scan rate of 10 mV s⁻¹. Reprinted from Ref. [188].

disadvantage for lithium batteries based on the liquid electrolytes viz. safety issue because of possible leakage. This also makes the cell design more expensive. All-solid-state cells have noticeable advantages but not appropriate for all applications. The biggest problem in solid electrolyte is the formation of highly resistive layers at the electrode/electrolyte interface [206,207]. The contact area between the electrode surface and solid electrolyte is very limited with less chance for self-repairing. This is the reason that it is crucial to soften the electrode/electrolyte interface in all-solid-state-cells [208].

A possible alternative to rigid solid electrolytes is gel polymer electrolytes providing ionic conductivity through a solid but flexible membrane [209–218]. Polymer gel electrolytes are composed of a polar polymer such as PEO, PEG, PVA, PMMA, PvdF-HFP, etc. and an ionic salt such as LiBF₄, LiClO₄, NH₄ClO₄, etc. Despite a considerable attention paid to this alternative during the past two decades, low ionic conductivity has made them impractical. Several methods have been introduced to improve the ionic conductivity of gel polymer electrolytes. The most common way is to add organic plasticizers such as EC, PC, DEC, etc., which are indeed the common problematic electrolytes of conventional lithium batteries. Subsequently, lithium batteries utilizing this class of gel polymer electrolytes still suffers from common safety issues such as flammability, toxicity, etc.

Ionic liquids can well address this issue, and thus, gel electrolytes made by dissolving ionic liquids within polymer matrix are now a promising candidate for the next generation lithium batteries [219–267]. Fig. 5 typically shows the flexibility of gel electrolyte, which is beneficial for easier casting. In this case, IL acts as both plasticizer and charge carrier. In fact, ILs are the biggest hope to

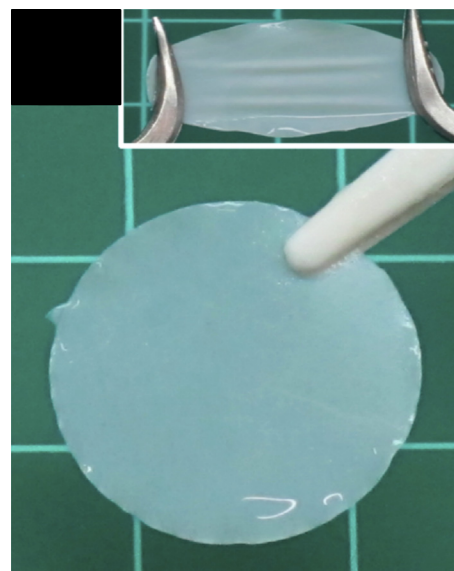


Fig. 5. Typical gel polymer electrolytes. The insets show the flexibility of the membranes. Reprinted from Ref. [267].

make gel polymer electrolytes practical [268]. Upon reaching this advancement, the applications are not limited to lithium batteries, as IL-based polymer gel electrolytes can be the next generation of ionic membranes. For example, the central issue of lithium-air batteries is to have an ideal membrane separating metallic

lithium from air, while Li diffusion is facile; and IL-based polymer gel electrolytes have shown promising advancement in this direction [244].

Since this is a gel matrix, diffusion of Li is through the gel channels rather than liquid medium; thus, the low viscosity of ILs is not a critical issue here. While the gel electrolyte acts as the cell separator too, its flexibility provides an excellent opportunity for casting and cell design. In general, gel electrolytes have noticeable advantages in comparison with solid electrolytes or pure IL electrolytes; however, they still have some disadvantages in comparison with conventional organic electrolytes such as slower diffusion and mismatch at the electrode/electrolyte interface.

Interestingly, gel electrolytes can be prepared merely by ILs without any polymer [269]. In this design, an IL acts as the solid matrix while the other IL is the plasticizer. The capability of ILs for forming gel materials has been extensively reviewed by Marr and Marr [270].

6. Ionic liquids as binders

Application of ILs for lithium batteries is not limited to electrolyte only. Similar to immobilization of ILs within the polymer matrix to form a gel electrolyte, ionic forces of ILs can make links between electroactive particles [271–279]. Carbon materials are commonly used to improve the electrical conductivity of cathode materials (particularly less conductive materials such as LiFePO_4). To reach this goal, it is essential to make powerful bonds between the carbon additive and electroactive material. ILs are made of ions, which can be adsorbed on particle surfaces depending on the chemical composition. A working example is functionalizing the carbon surfaces in which the charged ions interact with free π electrons at the carbon surface; then, a gelatinous material is formed [278,279]. This functionality has paved the path to use ILs as binders for casting electrode materials [279].

Application of IL binders is not limited to the classical approach of mixing the electroactive material with the binder mechanically. ILs are like an octopus with lots of ions, which may have different functionalities. For instance, the free side of IL in IL-functionalized single wall carbon nanotubes (SWCNTs) can assist in unzipping the SWCNTs [277]. This is a reminder of the well-known scotch-tape method first used to separate graphene out of graphite structure [280]. IL not only wraps the separated carbon layers around the electroactive nanoparticles to coat them but also acts as a binder between the nanoparticle and carbon coating (Fig. 6a). An interesting feature of this structure is that the binding IL is not a solid binder, and thus, the Li ions are free to move along the IL layer.

This creates shortcuts for the diffusion pathway, as Li-ions do not need to diffuse toward the particle in perpendicular directions to shorten the solid-state diffusion length (Fig. 6b). Because of this feature, ILs are widely used as binders for carbon paste electrodes in electroanalytical chemistry [281–293]. Such experiences can be directly used in lithium batteries.

A new type of IL binders has been introduced by the development of poly(ionic liquid)s (PILs), which are usually ionic nanoparticles [294–302]. The potential of PILs as binders for electrode materials has not been well understood yet [272,273]. PILs can have all advantages of available polymeric binders plus an extraordinary power as they are electrochemically permeable (Fig. 7). Typical binders such as PVdF well improve the electrode mechanically and electronically, but at the cost of blocking the Li diffusion (Fig. 7c). PIL binders can enhance the specific capacity by making the electroactive material more accessible interfacially (Fig. 7a) while the cyclability is also improved. The latter is possibly because of non-blocking diffusion pathways through PIL binders, which avoid several structural changes as the result of Li interactions with the blocking PVdF binder.

This unique collection of features can lead to a new idea to use PILs as protective coatings too. It has been shown that when a coating is of the material utilized in the electrode composite, they can make intrinsic bonds improve mechanical stability and open channels for Li diffusion [303]. Along with conductive polymers, which are electrically conductive because of the free π electrons; PILs, which are ionically conductive, can be a new chapter in the realm of polymer science.

7. Prospective: supercooled ionic liquid crystals

As discussed above, a practical approach to gain the unique features of ILs while planning a fast diffusion is to conduct the diffusion process through channels more like a solid structure rather than liquid medium. Free movement of a liquid species provides an excellent opportunity for fast diffusion, but in the case of ILs, it is not that easy due to the friction of charged ions (causing high viscosity) and complicated interactions of the diffusion ions within the ion-based structure of ILs. Therefore, it can be a brilliant idea to build a solid-like structure within an IL to create spacious channels for diffusion without interfering interactions.

This approach does not need a discovery, as liquid crystals are well known for a century [304–313]. Although the primary attention to liquid crystals is related to optical properties, which have been used in commercial products for decades; there is a growing interest in electrochemical systems [314–334] including lithium

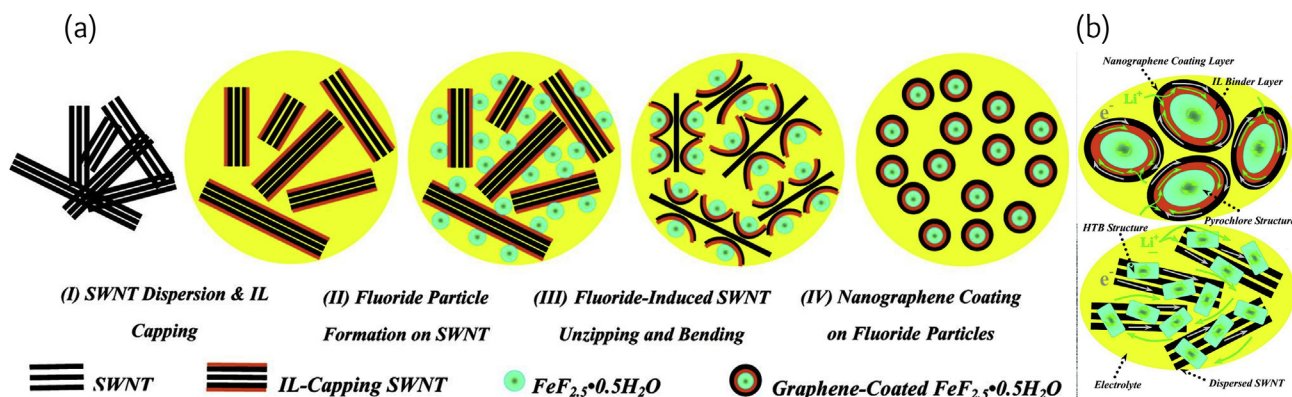


Fig. 6. (a) A schematic of functionalizing SWCNT surfaces and unzipping the carbon sheets while covering the nanoparticles. (b) A possible diffusion pathway through IL layer. Reprinted from Ref. [277].

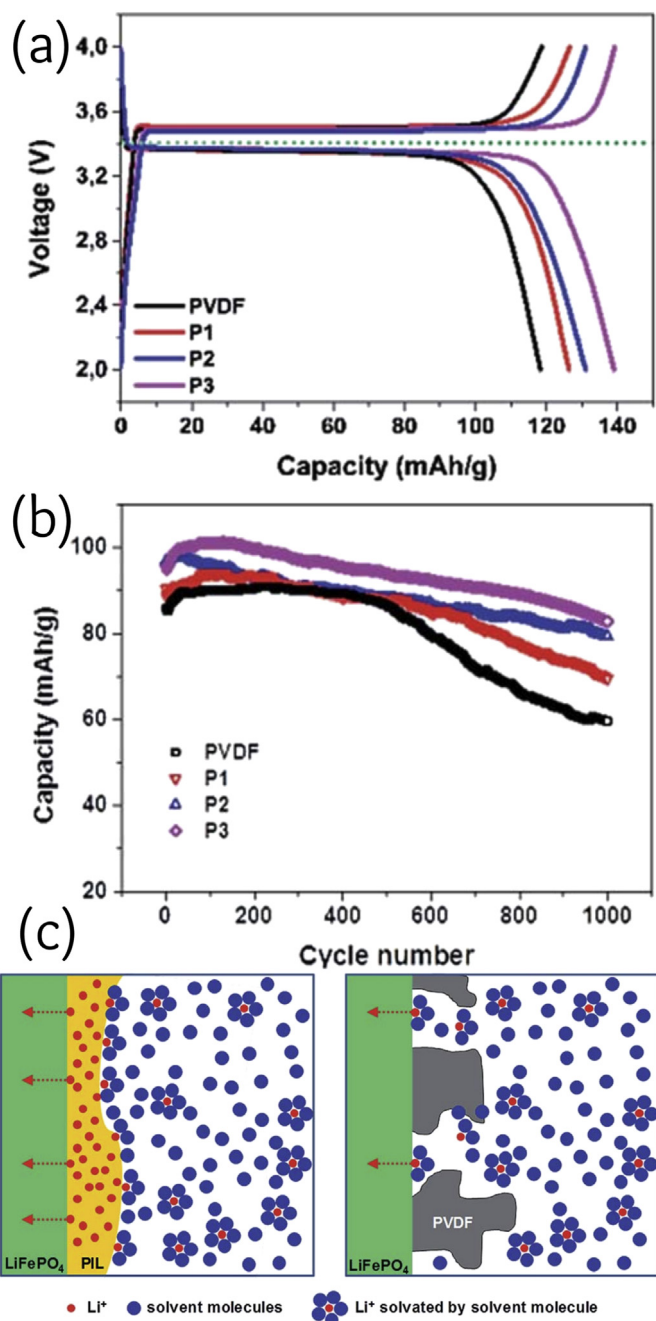


Fig. 7. (a) Charge-discharge profiles and (b) cyclability of a LiFePO_4 electrode bound by PIL and PVDF. (c) A schematic of a LiFePO_4 particle bound by PIL (left) and PVDF (right). Reprinted from Ref. [273].

batteries [335–337], because of the ordered species channels within liquid crystals, which are very appropriate for diffusion of electroactive species. Kato and his coworkers utilized the anisotropic nature of liquid crystals to obtain ionically functional electrolytes in a simple electrochemical cell design [334]. Diffusion of Li along the aligned molecules is significantly faster than that in randomly oriented molecules, due to lower activation energies [338]. However, liquid crystals cannot find a universal place in electrochemical systems as ILs did, because of conductivity issue. Fortunately, ionic liquid crystals (ILCs) are also known for decades (almost as old as ILs) [339–350].

ILCs are rapidly extending the applications of liquid crystals in electrochemical systems [351–357] by providing the possibility of

ionic conductivity. ILCs have been successfully utilized as the electrolytes in various electrochemical systems [354,358–362]. Diffusion of Li is usually slower than its counterpart anion due to the coordination of Li cations with oxygen-contained species in the electrolyte, but diffusion is significantly faster along the oriented structure of ILCs [177]. There is also a possibility for hopping mechanism. In general, the formation of an LC smectic layer provides secure pathways for ion mobility and thus ionic conductivity [177].

As presented in Fig. 8, long alkyl chains keep the diffusion paths separated. On the other hand, the vertical arrangement of these alkyl chains makes an ordered direction for all channels. Therefore, diffusion of Li ions can be one-directional through guided channels. If arranging these channels perpendicular to the electrode surface, the ILC electrolytes resemble diffusion through highly ordered microporous materials, but deep within the electrolyte medium.

Supercooling is a common phenomenon for ILs [363–372], but it is not of practical interest. The primary focus in IL studies is to reduce the viscosity but supercooling significantly increases the viscosity. However, in the case of ionic liquid crystals, the viscosity is not the key factor for diffusion but the diffusion pathways, which can be more ordered by supercooling.

It is expected to have a more ordered structure in supercooled ILs, but the dynamics of diffusion are very complicated. Diffusion in supercooled ILs does not obey the Stokes-Einstein, or Debye-Stokes-Einstein laws and diffusion can become faster by decreasing the temperature [208,373–376].

The idea of supercooled ionic liquid crystals shed light on the future of ILs for lithium batteries. The capabilities of ILs are much more than conventional molecular solvents. In other words, it is not the best choice to imitate the characteristic properties of conventional solvents (e.g., attempting to reduce the IL viscosity simply) to build ideal ILs as electrolytes of lithium batteries. Instead, a new generation of electrolytes and diffusion systems should be designed for the next generation of lithium batteries. Theoretically, a gel electrolyte made of supercooled ionic liquid crystals can be an ideal electrolyte far beyond available possibilities; however, not a

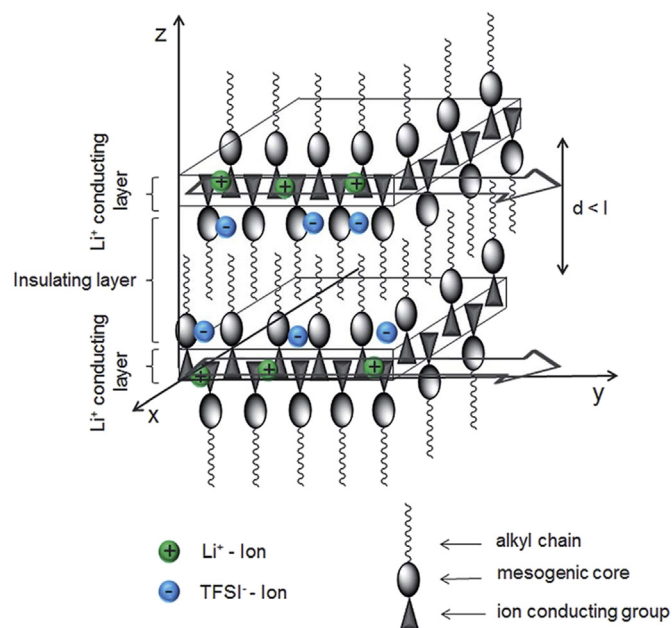


Fig. 8. Schematic illustration of anisotropic Li ion conductivity along the layer plane in x- and y-direction when the molecules align in an oriented interdigitated molecular SA phase. Reprinted from Ref. [177].

practical step has been taken in this direction, as the focus has been on reaching the specific features of commercial organic electrolytes.

In conclusion, ILs are no simple replacements for organic carbonates; ILs bring a broad range of new possibilities into consideration to design a new generation of electrolytes with entirely new sets of features and expectations.

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