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COMMUNICATION

A fluorine-substituted pyrrolidinium-based ionic liquid for high-voltage Li-ion battery

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A fluorine-substituted ionic liquid based on pyrrolidinium cation and bis(fluorosulfonyl)imide anion was synthesized using a facile one-step reaction. The resulted ionic liquid is highly pure and when dissolved with LiFSI, the IL-based electrolyte showed good compatibility both Li and graphite anodes, and superior voltage stability demonstrated in a LiNi_{0.5}Mn_{0.3}Co_{0.2}O₂ cell.

As demands in modern portable electronics and electric vehicles (EVs) increase, lithium-ion battery (LIB) has been the fast growing and most promising battery chemistry due to its many advantages over other battery chemistries.¹⁻⁵ However, to extend electric driving range of EVs, the energy density of next-generation LIB needs to be improved. Also, to ensure the reliability of the LIB as a daily commodity, the safety of next-generation LIB needs to be enhanced.^{4,6} To meet both requirements, development of a high-voltage and non-flammable electrolyte is crucial.⁷ The state-of-the-art carbonate-based electrolyte, which is designed for the 4-V LIB chemistry, decomposes beyond 4.3 V vs Li⁺/Li, which could not support the redox reaction of high-voltage high-capacity cathode occurring at potentials >4.3 V.⁸⁻¹¹ Moreover, there is severe safety concern of this highly flammable electrolyte due to the presence of highly volatile organic carbonate solvents.^{1,5}

In contrast to the traditional organic carbonates, ionic liquid (IL) has wide electrochemical window, non-flammability and extremely high thermal stability.¹²⁻¹⁶ Therefore, ionic liquids have been considered as a promising alternative electrolyte solvent and have attracted lots of interests worldwide. Although they have been reported to enable the chemistries of lithium batteries, ideally, they also have the potential to improve their performance, especially with respect to cycling stability at high voltages and high temperatures. Ionic liquids

consist of a large organic cation and a smaller inorganic/organic anion. Therefore, the physical and electrochemical properties could be systematically tuned by changing the structures of cations or anions.¹⁷⁻²⁰ For example, ionic liquids based on PF₆⁻ or BF₄⁻ anions have melting point greater than 60°C (e.g. PMpyr-PF₆ m.p. 113°C, PMpyr-BF₄ m.p. 64°C),²¹ which cannot be used as bulk solvent for room temperature LIB. When switching the anions to sulfonyl amide, the melting point of corresponding ionic liquids lowers to below room temperature (e.g. PMpyr-TFSI m.p. 12°C, PMpyr-FSI m.p. 5°C).²² Moreover, the viscosity of the sulfonyl imide anions based ILs are relatively less viscous due to their electron-delocalized structure. Therefore, they are the most widely studied anions in ionic liquids used in LIBs. Compared with bis(trifluoromethylsulfonyl)imide (TFSI⁻), the bis(fluorosulfonyl)imide (FSI⁻)-based ILs are more favored mainly due to two reasons. First, the size of -F is smaller than -CF₃ group, therefore the ionic liquids based on FSI⁻ anions are less viscous than those based on TFSI⁻ anion. For example, the *N*-methyl-*N*-propyl pyrrolidinium-FSI (PMpip-FSI) has an ambient viscosity of 40 mPa·s, and it increases to 61 mPa·s when the anion is switched to TFSI⁻.²³ Second, the FSI⁻-based ILs could form solid-electrolyte-interphase (SEI) on graphite anode, therefore they are highly compatible with graphite anode.²⁴

Cations structure also shows big impact on the physical and electrochemical properties of ionic liquids. 1-Alkyl-1-methylpyrrolidine based ionic liquids have shown increased viscosity along with expanded electrochemical window when the alkyl side chain becomes longer (e.g. butyl vs propyl; higher viscosity and better electrochemical stability for the butyl).²⁵ Hassoun *et al.* reported that the ether-functionalized pyrrolidinium Pyr₁₂₀₁⁺-TFSI⁻ (1-methoxyethyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide) electrolyte has consequently lower viscosity and higher conductivity than the BMpyr-TFSI (1-butyl-1-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide) electrolyte.²⁶ However, introducing ether functional group compromises the high voltage stability of the ionic liquids.²⁷ In

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this paper, we report the synthesis and their electrochemical properties of two new ILs with fluorine and fluorinated alkyl functionalized cations, *i.e.* 1-methyl-1-propyl-3-fluoropyrrolidinium bis(fluorosulfonyl)imide (PMpyr_F-FSI) and 1-methyl-1-(2,2,3,3,3-pentafluoropropyl)pyrrolidinium bis(fluorosulfonyl)imide (P_FMpyr-FSI). A facile one-step route was adopted for the material synthesis which affords IL with extremely high purity. When dissolved with LiFSI salt, the new IL-based electrolyte demonstrates high voltage stability revealed by the cell performance in LiNi_{0.5}Mn_{0.3}Co_{0.2}O₂ (NMC532) cathode lithium cells.

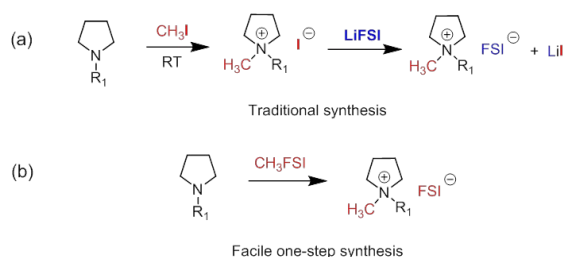


Fig. 1. (a) Representative synthesis of ionic liquids by traditional method and (b) facile one-step synthesis.

Traditional synthesis of ionic liquids includes two steps as shown in Fig. 1a:²⁸⁻³⁰ (1) Quaternization of a tertiary amine with alkyl halide to form quaternary ammonium salt and (2) anion metathesis of the quaternary ammonium salt with Li salt that contains target anions to form the target ionic liquid. For the Li-ion battery applications, solvent purity is critical for good cycling performance. This method introduces potential halide contaminations through the quaternization step.³¹⁻³³ Additionally, the purification at the end of the second step requires extensively aqueous wash/extraction and the determination of complete removal of halide impurities is based on the disappearance of precipitation formation when mixed with AgNO₃ solution, an arbitrary operation for high purity IL preparation. Furthermore, the availability and purity of the Li salt in the second metathesis step determine the accessibility and purity of the final ionic liquid. Here we report the new IL with a fluorinated cation using a facile one-step synthesis as generalized in Fig. 1b.³⁴ This method affords extremely high purity IL-based electrolytes which is pivotal for the long-term stability of LIB. Both precursors are organic compounds which could be synthesized and purified individually beforehand easily. When mixed together, the reaction occurs and forms the target IL with high purity. In this paper, two fluorinated pyrrolidines designed and synthesized and were treated with methyl bis(fluorosulfonyl)imide (CH₃FSI) to form the target ionic liquid in one-step. As the usage of alkyl halide was eliminated, no halide impurity exists.

1-Methyl-1-propyl-3-fluoropyrrolidinium bis(fluorosulfonyl)imide (PMpyr_F-FSI) and 1-methyl-1-(2,2,3,3,3-pentafluoropropyl)pyrrolidinium bis(fluorosulfonyl)imide (P_FMpyr-FSI) were successfully synthesized using the one-step synthesis (Fig. 2). 1-Boc-3-hydroxypyrrolidine was first reacted

with fluorinated reagent XtalFluoro-E^{35,36} to convert the hydroxyl group to fluoro group and the Boc-protecting group was then removed by HCl. The resulting 3-fluoropyrrolidine hydrochloride reacts with 1-bromopropane to form the 1-propyl-3-fluoropyrrolidine, which was purified by distillation. 1-Propyl-3-fluoropyrrolidine was then treated with CH₃FSI to form PMpyr_F-FSI (Fig. 2a) at room temperature with 100% conversion by NMR and 85% isolation yield. The -F group on the pyrrolidine ring backbone and methyl group on the pyrrolidine N atom can be on the same side or different side of the pyrrolidine ring. Correspondingly, two sets of pyrrolidine N-CH₃ group resonances and N-propyl group resonances are observed in ¹H NMR (Fig. S5a in SI). The ¹⁹F NMR (¹H decoupled) of PMpyr_F-FSI contains one resonance of FSI at δ 51.4 ppm and two resonances of -F on the pyrrolidine ring backbone at δ -168.9 ppm and -172.4 ppm (Fig. S5b in SI), respectively, indicating that PMpyr_F-FSI is formed as a mixture of diastereomers.

1-(2,2,3,3,3-Pentafluoropropyl)pyrrolidine was synthesized by reacting pyrrolidine with 2,2,3,3,3-pentafluoropropyl trifluoromethanesulfonate, where three equivalent of pyrrolidine was used to serve as reactant, solvent and quenching reagent for the triflic acid to form pyrrolidinium triflate.³⁷ The resulting triflate salts were then removed by filtration and the excess pyrrolidine was removed by HCl wash. The fluorinated pyrrolidine was purified by distillation and then treated with CH₃FSI to yield P_FMpyr-FSI with 83% isolation yield (Fig. 2b). The detailed synthesis and spectroscopic data are provided in the Supporting Information.

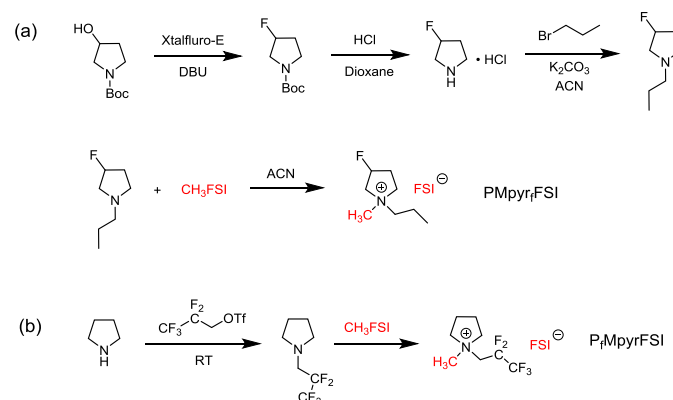


Fig. 2. (a) Synthesis of fluorine-substituted ionic liquid PMpyr_F-FSI and (b) P_FMpyr-FSI.

Electrochemical stability of the fluorinated ILs dissolved with 1 M LiFSI salt was measured by cyclic voltammetry. 1 M LiFSI PMpyr_F-FSI electrolyte shows a wide electrochemical window and high anodic stability up to 5.5 V vs Li⁺/Li (Fig. 3a), which is slightly higher than the none functionalized pristine IL electrolyte (about 5.4 V vs Li⁺/Li, Fig. S8b). Interestingly, a reductive peak is observed around 3 V during the cathodic scan which is correlated to the oxidative decomposition product generated at potential > 5.5 V during the anodic scan. As the scan upper voltage was limited to 5 V, no such a reduction peak was observed. The oxidation peak around 0.5 V

and reduction peak below 0 V are assigned to be Li stripping and plating peaks, respectively. Two more oxidation peaks that observed in the anodic scan around 0.7 V and 1.4 V are assigned to be the stripping peaks of the Li-Pt alloying.³⁸⁻⁴⁰ Table S1 summarized the ambient viscosity and conductivity of PMpyr_F-FSI, P_FMpyr-FSI and pristine PMpyr-FSI ionic liquid and the electrolytes. After introducing fluoride onto the cation structure, the viscosity shows dramatic difference. The single fluorine substitution on the cyclic ring leads to a lower viscosity of 83 mPa.s, where the fluorinated side alkyl chain substitution significantly increases the viscosity (261 mPa.s). As expected, the conductivity for PMpyr_F-FSI is higher than P_FMpyr-FSI. Furthermore, the conductivity of PMpyr_F-FSI and P_FMpyr-FSI decrease and viscosity increase with the increased LiFSI salt concentration, which is consistent with previous literature on the impact of salt concentration on the conductivity and viscosity of ionic liquids.^{41,42} Specifically, for PMpyr_F-FSI, with 1 M LiFSI, the conductivity decreases nearly half to 2.58 mS cm⁻¹ and viscosity nearly doubles to 150 mPa.s at 25°C. When the LiFSI salt concentration increases to 4 M, the ionic conductivity further decreases to 0.55 mS cm⁻¹ and viscosity increases to 528 mPa.s. Due to the intrinsic high viscosity of P_FMpyr-FSI, with 4 M LiFSI salt concentration, the viscosity increases to over 1000 mPa.s and conductivity drops to 0.36 mS cm⁻¹.

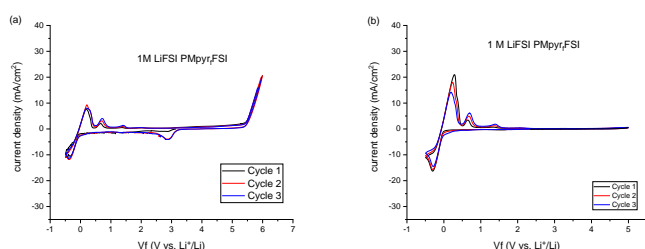


Fig. 3. Cyclic voltammetry profiles of 1 M LiFSI PMpyr_F-FSI. (a) Scan potential vs Li⁺/Li from -0.5 to 6 V, and (b) from -0.5 to 5.0 V using a Pt/Li/Li three electrochemical cell (scan rate 10 mVs⁻¹).

Fig. 4a shows the cycling performance of 1 M LiFSI PMpyr_F-FSI electrolyte and 4 M LiFSI PMpyr_F-FSI electrolyte in a NMC532/Li cell with a cutoff voltage of 4.3-3.0 V. To avoid the corrosion reaction of stainless-steel, aluminum-coated coin cells are used for all the electrochemical testing. The cells were formed with C/20 for 3 cycles followed by 100 cycles at C/10 rate. 145 mAhg⁻¹ initial specific discharge capacity and 147 mAhg⁻¹ at cycle 100 were obtained for 1 M LiFSI PMpyr_F-FSI electrolyte cell. The reasons for the slightly increased capacity were two-fold: (1) PMpyr_F-FSI has high electrochemical stability resulting in nearly no capacity fading in the first 100 cycles, and (2) the intrinsic high viscosity of the ionic liquid causes a slow wetting process. As super-concentrated IL has shown improved cyclability and rate capability compared to dilute system in previous studies,⁴³⁻⁴⁵ we further test the ionic liquids with increased LiFSI salt concentration. For 4 M LiFSI PMpyr_F-FSI, the initial specific discharge capacity slightly increases to 148 mAhg⁻¹, and the slow wetting was observed for this electrolyte as the specific discharge capacity slowly

increases during cycling and reaches to 159 mAhg⁻¹ at cycle number of 100. The first cycle Coulombic efficiency for 1 M LiFSI PMpyr_F-FSI and 4 M LiFSI PMpyr_F-FSI are 82.2% and 83.7%, respectively. Encouraged by the excellent cycling performance with upper cutoff voltage of 4.3 V, we further evaluated this ionic liquid at high voltage condition using a 4.7 V upper cutoff voltage. As shown in Fig. 4b, initial specific discharge capacity is 206 mAhg⁻¹ and first cycle Coulombic efficiency is 88.6% for 1 M LiFSI PMpyr_F-FSI and the capacity retention at cycle 100 is 87%. With increased the salt concentration, the initial specific discharge capacity is 207 mAhg⁻¹ and first cycle Coulombic efficiency is 89.3% for 4 M LiFSI PMpyr_F-FSI. The capacity retention increases to 89% at cycle 100.

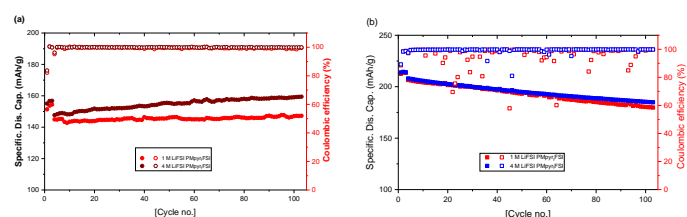


Fig. 4. (a) NMC532/Li cell cycling performance with a cutoff voltage 3.0-4.3 V and (b) 3.0-4.7 V.

PMpyr_F-FSI electrolyte was then evaluated in the NMC532/graphite full cells cycled between 3.0-4.2 V, with 3 formation cycles at C/20 and 100 cycles at C/10 rate. For 1 M LiFSI PMpyr_F-FSI, the first cycle specific discharge capacity is 96 mAhg⁻¹ with Coulombic efficiency only 51.3%, while for 4 M LiFSI PMpyr_F-FSI, the first cycle specific discharge capacity is increased to 142.7 mAhg⁻¹ and Coulombic efficiency increases to 78.4% as shown in Fig. 5. Furthermore, the capacity decreases rapidly for 1 M LiFSI PMpyr_F-FSI cycle by cycle, and the Coulombic efficiency, although slowly increases from the 1st cycle, is only below 99.5% during majority of cycles. These results suggest that the SEI formed on the graphite anode is more resistive in 1 M LiFSI PMpyr_F-FSI, therefore Li plating might occur during cycling causing low Coulombic efficiency. However with increased LiFSI salt concentration, regardless of its low ionic conductivity, a more conductive SEI forms,^{43,45,46} which facilitates the Li⁺ transport during cycling and up to 99.9% Coulombic efficiency is obtained.

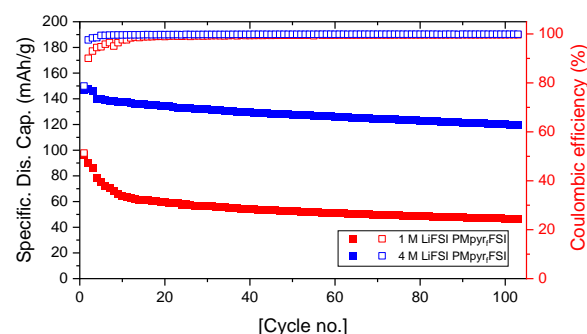


Fig. 5. Cycling performance of NMC532/graphite cells with 1 M and 4 M LiFSI PMpyr_F-FSI cycled between 3.0-4.2 V.

In sum, by reacting fluorinated alkyl pyrrolidine with CH₃FSI, the target ionic liquid 1-methyl-1-propyl-3-fluoropyrrolidinium bis(fluorosulfonyl)imide (PMpyr_F-FSI) and 1-methyl-1-(2,2,3,3,3-pentafluoropropyl)pyrrolidinium bis(fluorosulfonyl)imide (P_FMpyr-FSI) were successfully synthesized in a facile one-step reaction. The new reaction completely eliminates the halide contamination existing in the traditional anion metathesis method. The synthesized PMpyr_F-FSI shows high oxidation stability (> 5.5 V vs Li⁺/Li) as revealed by the cyclic voltammetry and good high voltage cyclability up to 4.7 V in NMC532/graphite full cells. Further formulation with increased LiFSI salt concentration to 4 M has shown increased deliverable capacity and cycling stability in both NMC532/Li and NMCC532/graphite cells. This simplified synthesis for functional ionic liquid opens up the opportunities for large scale, extremely high purity, and low cost IL-based electrolytes for safe electric vehicle application.

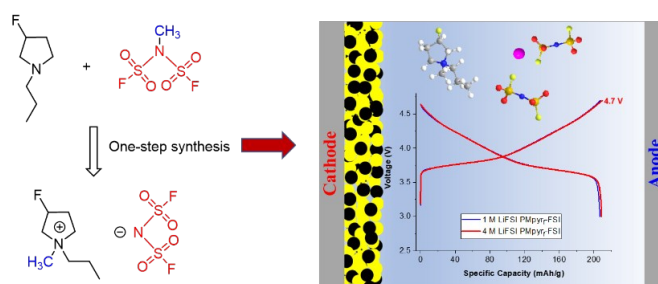
Conflicts of interest

There are no conflicts to declare.

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Fluorinated pyrrolidinium bis(fluorosulfonyl)imides were successfully synthesized by a facile one-step reaction by reacting fluorinated alkyl pyrrolidine with CH_3FSI with high purity and yield. The new electrolyte showed good performance in Li-ion battery.