

# Liquid Electrolytes Based on Ionic Liquids for Lithium-Ion Batteries

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**Abstract** Low-melting temperature ionic liquids (ILs), based on aliphatic quaternary ammonium  $N_{1xxx}$  and 1-ethyl-3-methylimidazolium cations with the imide anion, were prepared and characterized. The physicochemical and electrochemical properties of these ILs, including melting point, degradation temperature, viscosity, density, ionic conductivity and electrochemical stability, were determined. The fluorinated imidazolium cation was also studied because of its better electrochemical stability compared to the non-fluorinated form. Due to the high viscosity of aliphatic ammonium based ILs, addition of an organic solvent to the ionic liquid was investigated and found to be without detriment to the interesting properties of ILs (i.e., nonvolatility, non-flammability, high thermal stability, electrochemical stability). The cycling performance of the mixtures IL–ethylene carbonate was characterized using LiMn<sub>2</sub>O<sub>4</sub> based electrode materials in the voltage range of 3.5–4.3 V.

Keywords Ionic liquid · Electrochemical properties · Lithium battery · LiTFSI

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# 1 Introduction

Organic solvents are widely used in commercial lithium ion batteries with electrolytes having reasonable electrical conductivity and good compatibility with electrode materials. Nevertheless, the reactivity and volatility of organic electrolytes are disadvantageous to commercial applications [1]. In addition, they are all oxidized at potential over 4.5 V versus  $\text{Li}^+/\text{Li}$ , which can't be used for high voltage electrode materials such as  $\text{LiNi}_{0.5}$  Mn<sub>1.5</sub>O<sub>4</sub> which have working potential up to 5 V versus  $\text{Li}^+/\text{Li}$  [2]. The use of ILs in lithium ion batteries has been investigated for more than one decade [3] due to their interesting properties, such as non-volatility, non-flammability, high thermal stability, wide liquid range and wide electrochemical window. In addition, the choice of cation and anion can be made to tailor the properties such as ionic conductivity, viscosity and electrochemical stability [4–6].

The most promising of ILs used in Li ion batteries are based on the anion bis(trifluorormethanesulfonyl)imide, TFSI<sup>-</sup>, combined with different cations such as: quaternary ammonium, imidazolium, pyrrolidinium, and piperidinium. Due to the high flexibility, low geometric symmetry and good electrochemical stability of the TFSI<sup>-</sup> anion, these ILs exhibit low-melting points and low viscosities. Imidazolium based ILs have low viscosities which seems to make them promising electrolytes in lithium batteries; however, the electrochemical stability seems to be insufficient for high voltage batteries (about 5 V vs. Li<sup>+</sup>/Li) [7, 8]. The quaternary ammonium based ILs exhibit relatively better electrochemical stability toward lithium metal but the high viscosity is still a big challenge. As a result, tailoring of the cation structure has been a recent key development in ionic liquid application. Some different approaches were proposed, such as variation of the alkyl chain length, addition of functional group (methoxy, nitrile,...) or using the  $-CF_3$  group to replace the  $-CH_3$  group of alkyl chains [5, 6]. Furthermore, the high viscosity of ILs can be also decreased by adding a small amount of polar organic solvent (e.g., ethylene carbonate, dimethyl carbonate) without negative effects on dissociation of the ILs and electrochemical stability [9, 10].

In this work, we studied the physicochemical and electrochemical properties of ILs based on aliphatic quaternary ammonium  $N_{1xxx}$  and 1-ethyl-3-methylimidazolium cations. Replacing the  $-CH_3$  group with the  $-CF_3$  moiety was studied for theimidazolium cation based ILs in order to compare with the results obtained with those of fluorinated quaternary ammonium or cyclic ammonium cations [5, 6]. We also continue to further explore the electrochemical properties and cycling behavior of the of IL–organic solvent mixtures (e.g., ethylene carbonate) at different amounts of solvent (vol.%) according to our previous results [11].

# 2 Experimental Section

#### 2.1 Synthesis of Quaternary Ammonium Based ILs

Alkylammonium bis(trifluorormethanesulfonyl) imide ionic liquids were synthesized by the metathesis reaction of freshly prepared halide salts of substituted ammonium with different alkyl chains, as previously reported [5].

The ethylene carbonate (EC) (Aldrich) was stored in molecular sieves in glove box. The mixtures of ILs–EC were prepared, with stirring, in the glovebox.

## 2.2 Synthesis of Imidazolium Based ILs

The ionic liquid EMI<sup>+</sup>TFSI<sup>-</sup> was synthesized by a one step methathesis: 1-ethyl-3methylimidazoliumchloride EMI<sup>+</sup>Cl<sup>-</sup> (1.465 g, 0.01 mol) and lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) (2.871 g, 0.01 mol) were dissolved in acetonitrile in two separate vials. An anion-exchange reaction occurred after adding slowly (drop by drop) LiTFSI solution in a 10 mL round-bottom flask containing the EMI<sup>+</sup>Cl<sup>-</sup> solution, whereby the mixture was precipitated. Then, the reaction mixture was stirred at 500 rpm at room temperature for 48 h. After removal of the solvent, the mixture was washed repeatedly with water until the Cl<sup>-</sup> could not be detected by addition of AgNO<sub>3</sub> solution. The organic phase was collected in a vial and was passed at least twice through Celite silica column with ethyl acetate to completely remove Cl<sup>-</sup>. After removal of the solvent, the final product was dried under vacuum to give a yellowish liquid (2.347 g, 62 %).

The fluorinated imidazolium based IL, (2,2,2-trifluoroethyl)-3-methylimidazolium bis(trifluoromethanesulfonyl)imide, was prepared through the tosylate derivative pathway without solvent and hazardous catalysts [12].

Water contents of all the ILs, determined by Karl-Fisher titration, were below 20 ppm. The ILs were stored in a glove box ( $[H_2O] < 3$  ppm).

## 2.3 Measurements

The structure of intermediate and final products were confirmed by <sup>1</sup>H-NMR and <sup>13</sup>C-NMR spectra measured on a Bruker 500 spectrometer (500 MHz for <sup>1</sup>H and 125 MHz for <sup>13</sup>C) in DMSO-d6 or acetone-d6, and mass spectra were recorded via GC–MS and LC–HR-MS techniques. GC–MS analyses were performed on an Agilent GC System 7890A, equipped with a mass selective detector Agilent 5973 and a capillary column HP-5MS (30 m × 0.25 mm × 0.25  $\mu$ m). LC-HRMS analyses were performed on a Bruker micrOTOF-Q II.

DSC measurements were performed using a Mettler Toledo DSC1 Star. Samples of 10 mg were sealed in aluminium pans in a glovebox. Each sample was heated from -80 to 100 °C at a heating rate of 10 °C·min<sup>-1</sup>. The temperature calibration was carried out following the standard procedure using pure metal weighed into the hermitically sealed aluminium crucible. The precision of this calibration is 0.5 °C for a scanning rate of 10 °C·min<sup>-1</sup>.

Thermogravimetric measurements were carried out with a TGA Q500 V20.10 Build. This instrument possesses a relatively automated mass calibration procedure. Temperature calibrations were performed by using the Curie points of standard metals. A few milligrams of the sample was heated from the room temperature up to 600 °C at 10 °C·min<sup>-1</sup> under nitrogen flow. The precision of this calibration is 0.5 °C for a scanning rate of 10 °C·min<sup>-1</sup>.

The ionic conductivities and viscosities of the ILs in the glove box were measured by using an OAKTON CON 2700 Conductimeter and a Cannon Ostwald viscometer.

Cyclic voltammetry of pure ILs was carried out at the a scan rate of  $1 \text{ mV} \cdot \text{s}^{-1}$  on a MGP2 Biologic instrument (France) using a three-electrode cell. The counter electrode was a Pt wire and the working electrode was a Pt electrode with diameters of 1 mm and 25  $\mu$ m, respectively. The reference electrode was a Ag wire in 10 mmol·L<sup>-1</sup> AgNO<sub>3</sub> in acetoni-trile + 0.1 mol·L<sup>-1</sup> tetrabutylammonium perchlorate (TBAP). Preparation of electrolyte

solutions was carried out in a glovebox to protect the ionic liquids from absorbing water. The resulting potentials can be converted to the Li<sup>+</sup>/Li half cell by adding 3.548 V.

The oxidation stability of the lithium electrolyte containing pure ILs with LiTFSI or mixtures of ILs-EC were also studied by cyclic voltammetry (CV). The lithium salt concentration was 0.25 or 0.5 mol·L<sup>-1</sup> in order to obtain the optimized ionic conductivity and viscosity of the electrolyte.

All galvanostatic cycling were performed on the three-electrode Swagelok cell using lithium metal foil (Aldrich, battery grade) as anode, glass microfiber (Whatman, GF/D) as separator, and a positive electrode prepared by combining LiMn<sub>2</sub>O<sub>4</sub> (synthesized in laboratory), PTFE 60 wt% in water as binder (Aldrich) and carbon C65 (Timcal, Swiss). The electrode paste was prepared by mixing and laminating to the thin film electrode with 150  $\mu$ m thickness. The final electrode composition was 80 wt% active material, 15 wt% conducting carbon, and 5 wt% PTFE binder. As electrolyte, 0.25 mol·L<sup>-1</sup> LiTFSI dissolved in mixed solution EC–ILs was utilized.

## **3** Results and Discussion

#### 3.1 Quaternary Ammonium Based ILs (QAILs)

The QAILs obtained and their properties are summarized in Table 1. Abbreviations for the substituted ammoniums ion were selected by subscripting the number of carbons in each of the four alkyl substituents. For aliphatic quaternary ILs, using a low symmetry cation of large size cation combined with low symmetry and relative large sized anion gives generally low melting temperature ( $T_m$ ). For the N<sub>112x</sub> series,  $T_m$  decreases with the alkyl chain length. A similar tendency was observed for N<sub>122x</sub> and N<sub>133x</sub>. The N<sub>1125</sub>, which has lower

Ionic liquids	T <sub>m</sub> (°C)	T <sub>d</sub> (°C)	Density (g·mL <sup>-1</sup> , 30 °C)	Viscosity $\eta$ (mPa·s, 35 °C)	Conductivity $\sigma$ (mS·cm <sup>-1</sup> , 35 °C)	Molar conductivity $\Lambda$ (mS·cm <sup>2</sup> ·mol <sup>-1</sup> , 35 °C)
N <sub>1118</sub> TFSI	7.0	380.0	1.26	145	1.10	0.39
N <sub>1116</sub> TFSI	32.1	395.2	1.32	119	1.70	0.54
N <sub>1114</sub> TFSI	8.3/17.0	400.3	1.41	88	3.30	0.95
N <sub>1233</sub> TFSI	15.0/29.0	405.0	1.32	84	2.10	0.63
N <sub>1123</sub> TFSI	-12.0	452.1	1.22	55	3.30	1.09
N <sub>1124</sub> TFSI	-10.0	446.0	1.39	70	2.60	0.77
N <sub>1125</sub> TFSI	-8.0	403.3	1.56	95	1.90	0.52
N <sub>1224</sub> TFSI	10.2	446.2	1.35	85	2.80	0.88
N <sub>1224f</sub> TFSI	36.0	420.0	-	361	0.50	0.16
N <sub>1225</sub> TFSI	-	445.2	1.31	88	1.80	0.60
N <sub>1334</sub> TFSI	-	485.1	1.23	289	0.70	0.26
N <sub>1335</sub> TFSI	-	433.3	1.20	269	0.70	0.27

**Table 1** Properties of quaternary ammonium imide ionic liquids

symmetry but the same molecular weight as  $N_{1224}$ , exhibited the low  $T_m$  of -8 °C, compared to -10 °C. Similar results were reported for  $N_{1123}$ TFSI and  $N_{1114}$ TFSI [5].

All the ILs exhibited excellent thermal stability up to 485 °C (Table 1). The degradation temperature ( $T_d$ ) decreases quickly with increasing number of carbons in the alkyl chain. The  $T_d$  of N<sub>1125</sub>TFSI is lower than for N<sub>1224</sub>TFSI due to the longer C<sub>5</sub> alkyl chain.

The density of QAILs, *d*, measured at 30 °C, lies in the range of  $1.22-1.50 \text{ g}\cdot\text{mL}^{-1}$ . It was observed that the least symmetrcal cation,  $N_{112x}$ TFSI, exhibited an increase in density as the alkyl chain length increases. However, in the case of  $N_{122x}$  or  $N_{133x}$ , the density tends to decrease weakly.

The ionic conductivities of the ILs are mainly governed by their viscosities and formula weight (i.e., the molar concentration). The evolution of conductivity with temperature is plotted in Fig. 1. The ionic conductivities decrease with the increasing alkyl chain length. The highest conductivity was obtained for  $N_{1123}$ TFSI. For compounds of the same moecular weight, particularly  $N_{1125}$  and  $N_{1224}$ TFSI, there are significant differences in conductivity. When the temperature increases, the IL's conductivity increases rapidly. As compared to the conductivity of the commercial electrolyte LiPF<sub>6</sub>/EC:DMC (1:1), the highest value of  $N_{1123}$ TFSI is still smaller at 30 °C, 3.3 mS·cm<sup>-1</sup> versus 5.0 mS·cm<sup>-1</sup>.

In order to compare the conductivity of the ILs at the same molar concentration, the molar conductivity was determined at 35 °C (Fig. 2). Except for the fluorinated ammonium IL, the molar conductivity increases with the molar concentration. A quasi-linear behavior is observed. This behavior may be explained by an increase in the viscosity with the increase in the molar mass (decrease in the molar concentration) and an increase in ion association, which may be related to the formation of ionic clusters, being more favorable for the ionic liquids with greater alkyl chain length. Moreover, the increase in alkyl chain length induces a decrease in the amount of dissociation. The N<sub>1224f</sub>TFSI exhibits the lowest conductivity, which may be due to its high viscosity associated with its high  $T_g$  value.



Fig. 1 Ionic conductivity as a function of temperature for several ionic liquids



Fig. 2 Molar conductivity as a function of molar concentration at 35 °C

To evaluate the impact of these two effects (viscosity and dissociation) on the ionic conductivity, the viscosities of the ILs were measured between -10 and 70 °C. The evolution of the viscosity is plotted versus  $T^{-1}$  (Fig. 3); the data obtained at 35 °C are given in Table 1 whereas the remaining values are reported in Table 2.



Fig. 3 Viscosity of various ionic liquids versus temperature

	-		-		-		
<i>T</i> (°C)	1000 K/ T	N <sub>1116</sub> TFSI	N <sub>1118</sub> TFSI	N <sub>1224</sub> TFSI	N <sub>1123</sub> TFSI	N <sub>1114</sub> TFSI	N <sub>1124f</sub> TFSI
-10.00	3.8023		2571				
-5.00	3.7313	1220		930.7	374.3	730.4	
0.00	3.6630	800.0	1039	615.3	252.9	507.2	4798.2
5.000	3.5971	545.0		424.1	177.8	356.8	2872.8
10.00	3.5336	385.9	498	301.4	134.9	262.1	1805.6
20.00	3.4130	204.5	257	160.7	82.2	147.8	773.4
30.00	3.3003	119.6	145	91.7	74.7	88.0	361.1
40.00	3.1949	80.0	92	59.8	70.5	59.2	199.8
50.00	3.0960	52.8	65	41.4	65.6	30.6	119.1
60.00	3.0030			29.7	60.6	26.5	22.0
70.00	2.9155			22.1		23.9	

Table 2 Viscosity values of linear ionic liquids as a function of temperature

Units of viscosity: mPa·s

The viscosity increases with an increase in the molar mass, as expected due to van der Waals interactions. For a given molecular weight, the viscosity increases with the length of the alkyl chain and as the symmetry of the cation increases. For example,  $N_{1116}$ TFSI exhibits a viscosity of 119 mPa·s at 35 °C, as compared to 85 and 84 mPa·s for  $N_{1233}$ TFSI and  $N_{1224}$ TFSI, respectively. As expected, the ILs' viscosities decrease with an increase in temperature. This effect is attributed to the increase in Brownian motion of the constituent ions of the ionic liquids (Fig. 3).

The electrochemical windows of pure ILs were determined by cyclic voltammetry on a platinum microelectrode. The voltammograms of ILs are given in Fig. 2. The anodic and cathodic limits ( $E_{anodic}$  and  $E_{cathodic}$ ) versus Li<sup>+</sup>/Li were arbitrarily defined as the potential at which the current density reached 3 times the background current. All the ILs present good electrochemical stability for oxidation, with oxidation potentials higher than 5.5 V versus Li<sup>+</sup>/Li. As reported by Phung et al. [5], the oxidation stability depends on the length of the longest alkyl chain of the ILs. The shorter it is, the higher is the oxidation potential. The values of oxidation stability were determined as: N<sub>1335</sub>TFSI (5.7 V vs. Li<sup>+</sup>/Li) < N<sub>1225</sub>TFSI (5.8 V) < N<sub>1125</sub>TFSI ~ N<sub>1224</sub>TFSI (6.0 V) < N<sub>1123</sub>TFSI ~ N<sub>1124</sub>TFSI (6.2 V). Compared to the commercial electrolyte (1 mol·L<sup>-1</sup> LiPF<sub>6</sub>/EC:DMC 1:1), the  $E_{anodic}$  value of most stable ILs is similar, but the oxidation current is much smaller than for the last one.

#### 3.2 Mixtures of ILs–Organic Solvent

The most symmetrical of these ILs,  $N_{1123}$ TFSI, exhibits higher viscosity in comparison to the conventional electrolyte (55.7 vs. 3.10 mPa·s, respectively). One approach to decrease the viscosity of an IL is by using a small amount of organic solvent that isn't detrimental to the high oxidation stability of ILs. Phung et al. [11] studied the addition of carbonate solvents to the aliphatic ammonium based ILs. The addition of a small amount of solvent, i.e. 20 vol.%. causes a notable increase in the conductivities. In addition, the oxidation

potential was reduced when a large amount of solvent was added. The potential was reduced by about 100 mV at 25 vol.%. EC in  $N_{1123}$ TFSI or in  $N_{1124}$ TFSI (Figs. 4, 5).

The addition of LiTFSI to the mixed electrolyte ILs–EC normally induces a decrease in conductivity that is then lower than observed in the corresponding pure ILs. The minium concentration of LiTFSI in the mixed electrolyte was 0.25 mol·L<sup>-1</sup> for battery testing. The electrolyte ILs–EC + 0.25 mol·L<sup>-1</sup> LiTFSI exhibits similar oxidation stability as the ILs–EC mixtures, particularly for N<sub>1123</sub>TFSI and N<sub>1124</sub>TFSI (Table 3).

The battery cycling test was performed using ILs + x vol.% EC + 0.25 mol·L<sup>-1</sup> LiTFSI (x = 20, 25 %) at the standard rate of C/10. The results seem to be promising for N<sub>1123</sub>TFSI + 20 vol.% EC + 0.25 mol·L<sup>-1</sup> LiTFSI compared to the commercial electrolyte 1 mol·L<sup>-1</sup> LiPF<sub>6</sub>/EC–DMC. In Fig. 6, the discharge capacity of 87–100 mAh·g<sup>-1</sup> for N<sub>1123</sub>TFSI + 20 vol.% EC + 0.25 mol·L<sup>-1</sup> LiTFSI has relatively good cycle efficiency. In Fig. 7, the N<sub>1124</sub>TFSI + 20 vol.% EC + 0.25 mol·L<sup>-1</sup> LiTFSI has relatively good cycle efficiency. In Fig. 7, the N<sub>1124</sub>TFSI + 20 vol.% EC + 0.25 mol·L<sup>-1</sup> LiTFSI exhibits lower discharge capacity of 65–75 mAh·g<sup>-1</sup> due to the high viscosity of the electrolyte solution ( $\eta = 13.3$  mPa·s).

### 3.3 Imidazolium Cation Based ILs

As reported earlier, the imidazolium based ILs exhibit lower viscosity than those with an aliphatic cation. The inconvenience of imidazolium based ILs is their insufficient electrochemical stability with anode materials due to the C-2 hydrogen's acidity. The oxidation feature can be improved by using a -CF<sub>3</sub> substituent in the cation structure. For example, by using [CF<sub>3</sub>CH<sub>2</sub>MIm][TFSI] to replace the [CH<sub>3</sub>CH<sub>2</sub>MIm][TFSI]. The physicochemical properties and conductivities of imidazolium based ILs are given in Table 4 in comparision to the lowest symmetry aliphatic ammonium based ILs.



Fig. 4 Cyclic voltammetry of different amounts of EC mixed in N<sub>1123</sub>TFSI



Fig. 5 Cyclic voltammetry of different amounts of EC mixed in N<sub>1124</sub>TFSI

<b>Table 3</b> Oxidation potentialimit of quaternay ammonium	Electrolyte	E versus Li <sup>+</sup> /Li/V
based ILs and mixtures of Ls–EC determined at	N <sub>1123</sub> TFSI	6.2
$= 10 \text{ mA} \cdot \text{cm}^2$	$N_{1123}TFSI + 20$ vol.% EC	6.2
	$N_{1123}$ TFSI + 25 vol.%. EC	5.9
	N <sub>1124</sub> TFSI	6.2
	$N_{1124}TFSI + 15 \text{ vol.\% EC}$	6.1
	$N_{1124}TFSI + 20 \text{ vol.}\% \text{ EC}$	6.2
	$N_{1124}$ TFSI + 25 vol.%. EC	5.9

The fluorinated imidazolium exhibits a lower melting temperature, about -22 °C instead of -17 °C for [CH<sub>3</sub>CH<sub>2</sub>MIm][TFSI]. The replacement of a -CH<sub>3</sub> group by a -CF<sub>3</sub> group induces a decrease in the IL's melting point; however, an increase of viscosity was observed. The viscosity of fluorinated imidazolium obtained is nearly that of N<sub>1123</sub>TFSI.

In Fig. 8 the oxidation stability of the fluorinated imidazolium is higher than that of the non-fluorinated one. Thus, it can be concluded that the incorporation of  $-CF_3$  moieties into the imidazolium structure significantly improve the IL's stability towards oxidation. The reduction wall of fluorinated imidazolium based ILs observed at 1.5 V versus Li<sup>+</sup>/Li is associated with the reduction of the imide anion [13]. The fluorinated imidazolium exhibits better oxidation stability, i.e. 5.7 V versus Li<sup>+</sup>/Li compared to 5.1 V versus Li<sup>+</sup>/Li for EMITFSI (non-fluorinated imidazolium). Similar results were reported for fluorinated quaternary ammonium cation based ILs and non-fluorinated ones [5]. These results are consistent with thoeritical calculation of HOMO values for the linear optimized geometries of the ILs. The incorporation of a fluorinated group induces a decrease in the HOMO



**Fig. 6** Galvanostatic cycling of  $N_{1123}$ TFSI + 20 vol.% EC + 0.25 mol·L<sup>-1</sup> LiTFSI at C/10 rate in the potential range 3.5–4.2 V, at room temperature



Fig. 7 Galvanostatic cycling of  $N_{1124}$ TFSI + vol.% EC + 0.25 mol·L<sup>-1</sup> LiTFSI at C/10 rate in the potential range 3.5–4.2 V, at room temperature

Table 4	Properties c	of imidazolium	based ILs in	comparison to al	iphatic quaterna	ry ammonium	based ILs
						-	

Ionic liquids	$T_{\rm m}$ (°C)	T <sub>g</sub> (°C)	T <sub>d</sub> (°C)	Density (g·mL <sup>-1</sup> , 30 °C)	Viscosity $\eta$ (mPa·s, 35 °C)	Molar conductivity $\Lambda$ (mS·cm <sup>-1</sup> ·mol <sup>-1</sup> , 35 °C)
[CH <sub>3</sub> CH <sub>2</sub> MIm][TFSI]	-17	-35	414	1.48	24.9	2.35
[CF <sub>3</sub> CH <sub>2</sub> MIm][TFSI]	-22/-12		417	1.54	55.1	0.54
N <sub>1123</sub> TFSI	-12		452	1.22	55.7	1.09



Fig. 8 Cyclic voltammetry of different ionic liquids based on fluorinated imidazolium and aliphatic ammonium cations

values, and thus it can be concluded that the fluorinated ILs are electrochemically more stable than the non-fluorinated one.

## 4 Conclusions

Aliphatic ammonium bis(trifluoromethane sulfonyl) imides have been prepared and characterized. The highest conductivity was obtained for the least symmetric aliphatic ammonium cation,  $N_{1123}$ TFSI; however the high viscosity is still a big challenge for using it as an electrolyte in lithium ion batteries. Using a small amount of ethylene carbonate solvent (EC), i.e. 20 vol.%, induces a significant decrease in viscosity and thus an increase in ionic conductivity. The galvanostatic cycling test showed the promising capacity value obtained for  $N_{1123}$ TFSI + 20 vol.% EC + 0.25 mol·L<sup>-1</sup> LiTFSI. The addition of the -CF<sub>3</sub> moiety replacing the -CH<sub>3</sub> group of the imidazolium cation showed an improvement of oxidation stability though it gives rise to a high viscosity for the ILs.

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