Fast cycling of Li/LiCoO$_2$ cell with low-viscosity ionic liquids based on bis(fluorosulfonyl)imide [FSI]$^-$

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Abstract

A charge–discharge cycling test of a Li/LiCoO$_2$ cell containing ionic liquids based on bis(fluorosulfonyl)imide ([FSI]$^-$) as the electrolyte media, revealed significantly better rate properties compared to those of cells using conventional ionic liquids. The use of an 1-ethyl-3-methylimidazolium (EMI$^+$) salt permitted the retention of 70% of the discharge capacity at a 4 C current rate. In contrast, similar performance of cells containing N-methyl-N-propylpyrrolidinium (Py$_{13}^+$) and N-methyl-N-propylpiperidinium (PP$_{13}^+$) salts of [FSI]$^-$ was limited to operation at 2 and 1 C current rates, respectively. However, the charge/discharge cycling stability of the cell with Py$_{13}$[FSI] was much better than that of the cell using EMI[FSI].
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1. Introduction

Room temperature ionic liquids (RTILs) possess suitable properties for safe lithium battery electrolyte media due to their non-volatility and non-flammability [1]. The few reports concerning lithium battery electrolytes refer only to RTILs based on 1-ethyl-3-methylimidazolium (EMI$^+$) without any additives [2,3]. However, the uses of RTILs in high-energy electrochemical devices, such as a 4 V class lithium battery, require consideration of both the electrochemical stability and viscosity of the RTILs. The cations studied most extensively, such as EMI$^+$, form low-viscosity RTILs. Unfortunately however, the electrochemical windows of these RTILs are limited not only by electrochemical reduction, but also by the oxidation of the EMI$^+$ [4].

Recently developed RTILs based on an aliphatic quaternary ammonium cation (AQA), such as trimethylpropylammonium (N$_{1113}^+$ or TMPA$^+$), N-methyl-N-butylpyrrolidinium (Py$_{14}^+$), N-methyl-N-propylpiperidinium (PP$_{13}^+$) coupled with bis(trifluoromethylsulfonyl)imide ([TFSI]$^-$), exhibit sufficiently wide electrochemical windows to allow the electrochemical deposition of lithium without any additives [5–10]. This characteristic leads to a good cycle performance of a high-energy lithium battery that employs a lithium negative electrode and a LiCoO$_2$ positive electrode without any additives [6,11–13]. However, the charge-discharge rate of these cells could not be increased over 1 C, since the RTILs based on AQA are more viscous than an EMI$^+$-based melt [3] and conventional molecular liquids [11]. Therefore, an improvement in the performance of lithium batteries with additive-free RTILs necessitates the development of low-viscosity RTILs based on AQA.

In this paper, we discuss how RTILs based on bis(fluorosulfonyl)imide ([FSI]$^-$) may be used as electrolytes for a Li/LiCoO$_2$ cell. These RTILs have been found to be some of the most attractive candidates for such low-viscosity RTILs [14,15]. Their physical properties, such as viscosity and conductivity, with and without added lithium salt (Li[TFSI]), were measured and compared with those of the corresponding RTILs based on [TFSI]$^-$. In addition, as a further comparison, we examined the charge–discharge characteristics of a

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Li/LiCoO$_2$ cell using the same experimental setup and the conditions stated in our previous reports [6,11].

2. Experimental

In this study, three RTILs based on [FSI]$^-$ were used as shown in Fig. 1. These highly pure RTILs (Dai-ichi Kogyo Seiyaku, EMI[FSI]: Elexcel IL-110, Py$_{13}$[FSI]: Elexcel IL-120, PP$_{13}$[FSI]: Elexcel IL-130) were dried under vacuum at 95 °C for 1 h. The residual water in the dried RTILs, which was measured using a Karl-Fisher moisture meter (Mitsubishi CA-07), was below 50 ppm.

The experimental conditions and apparatus used in this study have been described in previous papers [10,11]. The viscosity and conductivity of the [FSI]$^-$ melt were measured with a spindle viscometer (Brookfield model DV-III+, cone spindle CP-52) and a conductivity meter (Radiometer Analytical, model CDM230, frequency 23.4 KHz), respectively. Both measurements were performed in a dry chamber (DAIKIN, dew point < −50 °C). The polarization curves were evaluated with an electrochemical analyzer (ALS model 660) in an Ar-filled glove box (O$_2$ and water <5 ppm). An I$_3^-$/I$^-$ reference electrode was used that consisted of Pt wire immersed in EMI[TFSI] containing 0.060 mol dm^{-3} of [(n-C$_3$H$_7$)$_4$N]I and 0.015 mol dm^{-3} of I$_2$. The iodide redox solution in the reference electrode was separated from the measurement solution by a porous Vycol glass tip fitted onto the bottom of the reference electrode. The potential was referenced to the ferrocene (Fc)/ferrocenium (Fc$^+$) redox couple in each RTIL. For comparison, we conducted a charge–discharge (C–D) test of the Li/LiCoO$_2$ cell containing

the RTILs was using the same experimental setup and the conditions described in our previous reports [6,11]. A 0.3 mol kg^{-1} of Li[TFSI] (3 M, ca. 0.4 mol dm^{-3}) was used as the source of Li$^+$. A two-electrode cell sealed in an Al-laminate film was employed for the cycling test. The details of the laminate cell have been provided in our previous paper [6].

3. Results and discussion

3.1. Physical properties of RTILs based on [FSI]$^-$

The diffusion of Li$^+$ in the electrolyte media is an important factor controlling the charge–discharge (C–D) rate property of a lithium battery system. As shown in our previous reports [6,11], the C–D rate depends on the viscosity of the various RTILs employed. Since the RTILs have a viscosity two or three order of magnitude higher than that of conventional molecular liquids in general, a reduction in RTIL viscosity is essential for the development of a practical lithium battery that contains them.

Table 1 shows the conductivity and the viscosity of the RTILs based on [FSI]$^-$ compared with those based on [TFSI]$^-$, whose electrochemical stabilities have made them attractive candidates for a Li battery electrolyte. The viscosity of the [FSI]$^-$ melt was ca. 60% lower than that of the corresponding [TFSI]$^-$ melts with the same cation. Adding a certain amount of the lithium salt into the RTILs, required when using RTILs as lithium battery electrolytes, results in a significant increase of the viscosity of electrolyte produced [3]. Notably, however, as shown in Table 1, the [FSI]$^-$ melts did not become remarkably more viscous than the [TFSI]$^-$ melts, after the addition of the same amount of
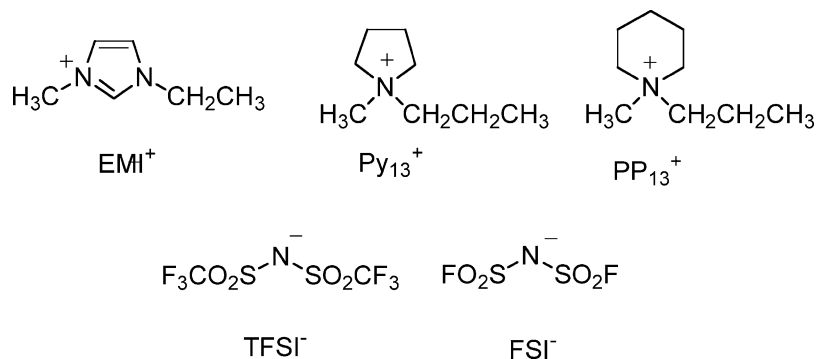

Fig. 1. Chemical structure of cations and anions containing ionic liquids electrochemically tested in this study: EMI$^+$, 1-ethyl-3-methylimidazolium; Py$_{13}$$^+$, N-methyl-N-propylpyrrolidinium; PP$_{13}$$^+$, N-methyl-N-propylpiperidinium; FSI$^-$, bis(fluorosulfonyl)imide; TFSI$^-$, bis(trifluoromethanesulfonyl)imide.

Table 1
Viscosity and conductivity of ionic liquids with and without 0.3 mol kg^{-1} of added Li[TFSI] at 298 K

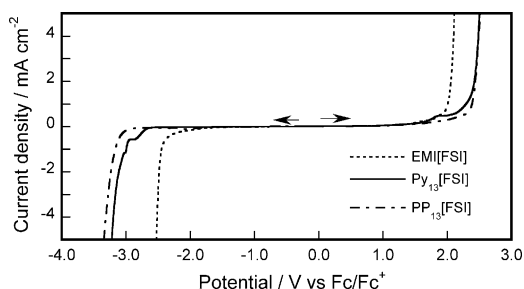|	Neat RTILs		With 0.3 mol kg^{-1} Li[TFSI]		Rate of increase	
	Viscosity (mPa s)	Conductivity (mS cm^{-1})	Viscosity (mPa s)	Conductivity (mS cm^{-1})	Viscosity	Conductivity
EMI[FSI]	18	15.4	24	11.0	1.3	0.71
Py$_{13}$[FSI]	40	8.2	51	5.5	1.3	0.67
PP$_{13}$[FSI]	95	3.7	124	2.5	1.3	0.68
EMI[TFSI]	33	8.3	55	5.1	1.7	0.61
Py$_{13}$[TFSI]	61	3.9	130	2.0	2.1	0.51
PP$_{13}$[TFSI]	151	1.4	330	0.73	2.2	0.52

Fig. 2. Linear sweep voltammogram of ionic liquids based on FSI$^-$ using a glassy carbon electrode: scan rate, 50 mV s^{-1}; temperature, 298 K; counter electrode, Pt wire. The potential (V) was referenced to the ferrocene (Fc)/ferrocenium (Fc$^+$) redox couple in each salt.

Li[TFSI]. As a result, the conductivity of the [FSI]$^-$ melts after the addition of 0.3 mol kg^{-1} of Li[TFSI] was maintained at ca. 70% of their neat melts, whereas for the [TFSI]$^-$ melts, it decreased over 50% from that of the neat melts. These results suggest that [FSI]$^-$ melts, as opposed to the [TFSI]$^-$ melts, will be very good RTILs for a lithium battery electrolyte.

3.2. Electrochemical windows of RTILs based on [FSI]$^-$

The electrochemical stability of the RTILs, which is often estimated on the basis of the electrochemical window (EW), is another important factor that governs such aspects of lithium battery performance as the output voltage [2,3], and the charge–discharge cycling property [6,11]. Fig. 2 shows a linear sweep voltammogram of the [FSI]$^-$ melts measured with a glassy carbon electrode. The cathodic or anodic limiting potential of an electrolyte was defined as occurring when the reduction or oxidation current reached a certain current density. Here, 1.0 mA cm^{-2} was selected as the cut-off current for comparison with our previous reports [10]. Table 2 shows the cathodic or anodic limiting potential of the various RTILs. The potential data in the table were referenced to the redox potential of ferrocene experimentally established in each melt.

For the EMI$^+$-based melt, a negative electrode such as Li[Li$_{1/3}$Ti$_{5/3}$]O$_4$, which has much more positive potential com-

Table 2
Electrochemical windows for various ionic liquids based on amide anions determined using a glassy carbon electrode at 298 K

Salts	Cathodic limit[a] (V)	Anodic limit[a] (V)	Electrochemical window (V)
PP$_{13}$[TFSI]	−3.4	2.5	5.9
PP$_{13}$[FSI]	−3.2	2.4	5.6
PP$_{13}$[TSAC]	−2.6	2.3	4.9
TMPA[TFSI]	−3.3	2.5	5.8
Py$_{13}$[FSI]	−3.0	2.3	5.3
EMI[TFSI]	−2.5	2.1	4.6
EMI[FSI]	−2.5	2.0	4.5

The potential was referenced to the redox potential of ferrocene in each of the ionic liquids. The data for PP$_{13}$[TFSI], PP$_{13}$[TSAC], TMPA[TFSI] and EMI[TFSI] was obtained from Fig. 2 of Ref. [10]. TFSI$^-$, [(CF$_3$SO$_2$)$_2$N]$^-$; TSAC$^-$, [(CF$_3$SO$_2$)N(COCF$_3$)]$^-$.

[a] The limit potential was defined as the potential at the current density when the linear sweep voltammogram reached 1 mA cm^{-2}.
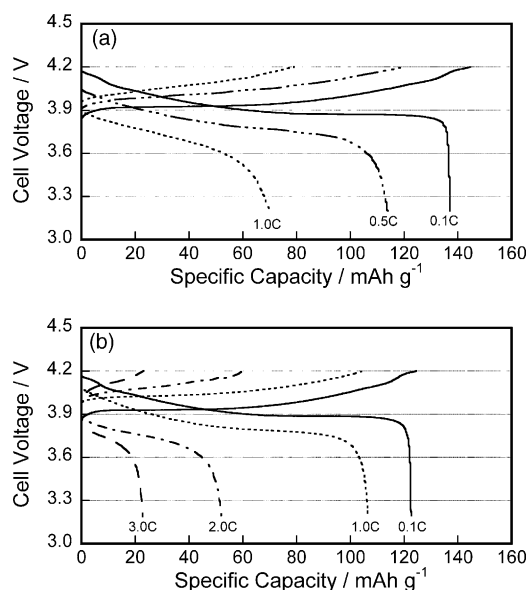
pared to a lithium metal or a graphite electrode, was necessary to achieve a good cycle stability [2,3]. The use of such negative electrode to avoid the decomposition of the EMI$^+$ resulted in about a 2.0 V class cell. Our previous study showed that the unavoidable use of a lithium metal electrode as a negative electrode in EMI[TFSI] resulted in a rapid decrease in the discharge capacity during the cycle testing of the Li/LiCoO$_2$ cell [11].

On the other hand, the reason why we have focused on RTILs consisting of [TFSI]$^-$ and an aliphatic quaternary ammonium (AQA), such as N-methyl-N-propylpiperidinium (PP$_{13}^+$) and trimethylpropylammonium (N$_{1113}^+$) is their wide EWs compared to EMI$^+$-based melts as shown in Table 2. This fact suggests that not only AQAs, such as PP$_{13}^+$ and N$_{1113}^+$, but also [TFSI]$^-$ is electrochemically more stable than the EMI$^+$ [10]. As shown in previous reports, a 4 V class cell (Li/LiCoO$_2$) could be constructed using PP$_{13}$[TFSI] [6,11]. In contrast, due to the poor electrochemical stability of [TSAC]$^-$ ([TSAC]$^-$: 2,2,2-trifluoro-N-(trifluoromethylsulfonyl)amide), the EWs of the RTILs based on that compound were quite narrow, despite its combination with PP$_{13}^+$ [10]. The degradation of the Li/LiCoO$_2$ cells consisting of the [TSAC]$^-$ melts was much more apparent than that of the [TFSI]$^-$ melt cells [11]. These observations strongly imply that not only the electrochemical stability of the cations, but also that of the anions might be important for constructing a 4 V class cell.

Based on the EW data of the RTILs with PP$_{13}^+$, the cathodic stability of the given three anions was in the order of [TSAC]$^- \ll$ [FSI]$^- \approx$ [TFSI]$^-$. On the other hand, the anodic stability of the anions was in the order of [TSAC]$^- <$ [FSI]$^- <$ [TFSI]$^-$. Thus, the performance of the cells with the [FSI]$^-$ melts is much higher than that of the cells with the corresponding [TFSI]$^-$ melts. However, as discussed in the next section, the slightly reduced anodic and cathodic stabilities of the [FSI]$^-$ melts compared with those of the [TFSI]$^-$ might be overcome by the formation of a surface passivation films on the electrodes, such as a solid electrolyte interface (SEI).

3.3. Charge and discharge (C–D) properties of Li/LiCoO$_2$ cell containing [FSI]$^-$ melt

All the components and experimental conditions used in the present study to construct a laminate-type cell, such as the type of lithium salt (Li[TFSI]) and the amount dissolved in the [FSI]$^-$ melts (0.3 mol kg^{-1}) were the same as those described in our previous reports [6,11]. First, to confirm the effect of the viscosity on the rate dependency of the Li/LiCoO$_2$ cell performance, we tested PP$_{13}$[FSI] and PP$_{13}$[TFSI], which show the highest electrochemical stability, but the highest viscosity for each anion. Fig. 3 shows the C–D curves for the RTILs based on PP$_{13}^+$ taken at various charge and discharge current rates. Although the initial capacity of the cell containing PP$_{13}$[FSI] (Fig. 3(b)) at 0.1 C was slightly smaller than that containing PP$_{13}$[TFSI] (Fig. 3(a)), the capacity ratio of 1/0.1 C (denoted as "capacity retention 1/0.1 C") of the PP$_{13}$[FSI] cell was more than 90%, a level much higher than that of the PP$_{13}$[TFSI] cell (50%). The difference in the initial capacity might have been caused by the wettability of the positive electrode in contact with the

Fig. 3. Charge–discharge curves of Li/Li[TFSI] cells with ionic liquids/LiCoO$_2$ at 298 K: (a) PP$_{13}$[TFSI]; (b) PP$_{13}$[FSI]. [Li[TFSI]] = 0.3 mol kg^{-1} (3.2–4.2 V, constant current mode). Charge/discharge rate is indicated in the figure.

electrolyte. In succeeding cycles, the PP$_{13}$[FSI] cell exhibited a sufficient capacity, and the wettability during the initial cycle was somewhat improved. These results clearly shows that the rate properties of the Li/LiCoO$_2$ cell could be enhanced using the less viscous RTILs, as we expected from the physical properties listed in Table 1. We also investigated the rate dependence of other [FSI]$^-$-based low-viscosity melts, such as Py$_{13}$[FSI] and EMI[FSI] (Fig. 4). In both cells, the initial capacity was adequate, and their rate properties further improved. For the Py$_{13}$[FSI] cell and EMI[FSI] cells, the capacity retention ratios 1/0.1 C were 87% and 93%, respectively. The EMI[FSI] cell could work consistently over many cycles even at the higher
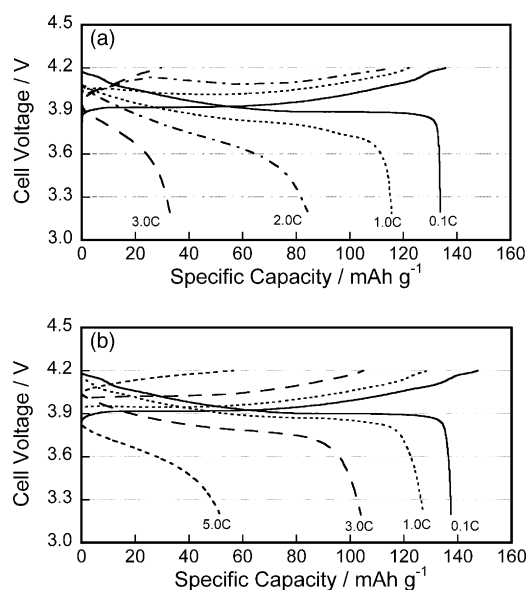

Fig. 4. Charge–discharge curves of Li/Li[TFSI] cells with ionic liquids/LiCoO$_2$ at 298 K: (a) Py$_{13}$[FSI]; (b) EMI[FSI]. [Li[TFSI]] = 0.3 mol kg^{-1} (3.2–4.2 V, constant current mode). Charge/discharge rate is indicated in the figure.
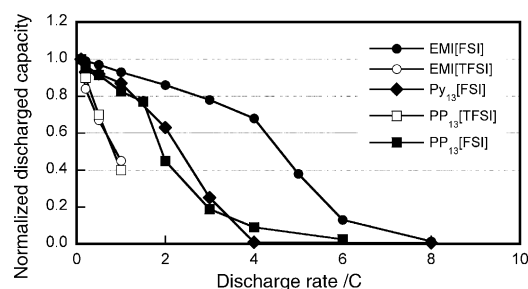

Fig. 5. Rate properties of Li/LiCoO$_2$ cell containing 0.3 mol kg^{-1} Li[TFSI] in various ionic liquids. The results for the TFSI melts were taken from Ref. [11]. Charge cut-off, 4.2 V CC mode; discharge cut-off, 3.2 V CC mode. Discharge capacity was normalized on the basis of the first discharge capacity at the 0.1 C rate of the same cell.

rate, and its capacity retention ratio 3/0.1 C was 75%. These results seemed quite reasonable, since the rate dependency of the Li/LiCoO$_2$ cell was thought to be governed by the diffusion of Li$^+$ in the RTILs. The rate dependencies of the cells containing several RTILs as the electrolyte are summarized in Fig. 5. The capacity of each cell at various current rates was normalized on the basis of the first discharge capacity at 0.1 C. One of the striking results in this study is that a rate over 4 C can be achieved by the use of EMI[FSI], which has a much narrower EW than that of PP$_{13}$[FSI] and Py$_{13}$[FSI] (Fig. 5). Furthermore, the reduced viscosity might not be the only reason for such a good cell performance with the [FSI]$^-$ melts. The rate dependence of the C–D property for the EMI[TFSI] electrolyte was quite inferior to that of the Py$_{13}$[FSI], which has a viscosity and conductivity comparable to that of EMI[TFSI] (Table 1). The difference between the cell performances of the EMI[TFSI] and EMI[FSI] reminded us that the rate dependence of a Li/LiCoO$_2$ cell depends not only on the diffusion of the Li$^+$, but also, as pointed out in our previous paper [11], on the interfacial reaction resistance of the redox reaction on the lithium metal electrode. Fig. 6 shows the cyclic voltammograms of the RTILs based on [FSI]$^-$ containing 0.3 mol kg^{-1} of Li[TFSI]. Even in the EMI[FSI] case, the plating and stripping peaks characteristic of the redox reaction of Li$^+$ on the Pt electrode could be observed, just as they were in the Py$_{13}$[FSI]. The redox potential of the Li$^+$/Li^0 couple in both the Py$_{13}$[FSI] and EMI[FSI] is about −3.2 V versus ferrocene (Fc)/ferricenium (Fc$^+$), a potential which is almost the same as that in the [TFSI]$^-$ melts such as PP$_{13}$[TFSI] and N$_{1113}$[TFSI] [10]. Though the addition of a Li salt improves the apparent cathodic limiting potential of EMI[TFSI], such a clear redox peak has never been seen in EMI[TFSI] under the same experimental conditions [16]. Therefore, these differences observed between the voltammograms of EMI[TFSI] and the EMI[FSI] indirectly suggests that the characteristics of the SEI film on an electrode in [FSI]$^-$ melts are much more favorable than those in [TFSI]$^-$ melts. Furthermore, as shown in Fig. 6, the anodic limiting potential of both of the RTILs based on [FSI]$^-$ was shifted about a few hundred mV toward the positive direction with the addition of Li[TFSI]. Considering the fact that such a potential shift in the anodic region was not observed for EMI[TFSI] or PP$_{13}$[TFSI] (not shown), we believe that the potential shift observed in [FSI]$^-$ melts might be caused by the generation of
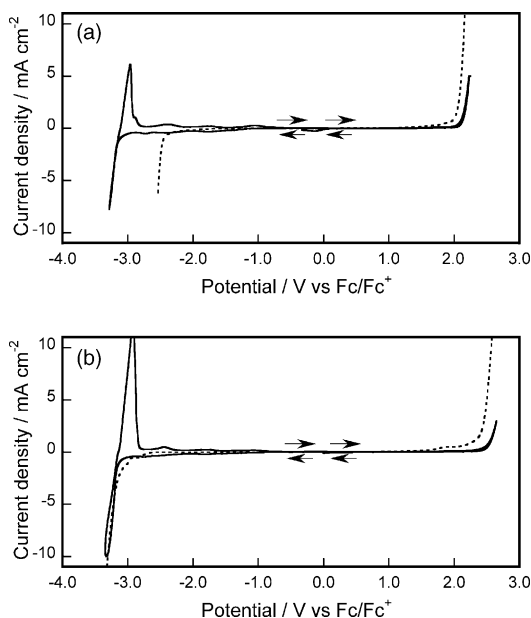
Fig. 6. Cyclic voltammogram of platinum electrode in RTILs based on $[FSI]^-$ containing 0.3 mol kg^{-1} of Li[TFSI]: (a) EMI[FSI]; (b) Py$_{13}$[FSI]. Temperature, 298 K; scan rate, 50 mV s^{-1}. The anodic and cathodic scans were separately measured with a fresh electrode. The linear sweep voltammogram of glassy carbon in neat $[FSI]^-$ melt is also indicated in each figure (dotted line).

a certain surface film (SEI) on the Pt electrode. The major component of the SEI film could be decomposed species from the $[FSI]^-$. To investigate the origin of the difference in the cell performance and the effect of the addition of Li salt observed in the voltammograms of the various RTILs with the same physical properties, we are now conducting a surface analysis study of the lithium negative electrode or a LiCoO$_2$ positive electrode by means of AC impedance spectroscopy and X-ray photoemission spectroscopy.
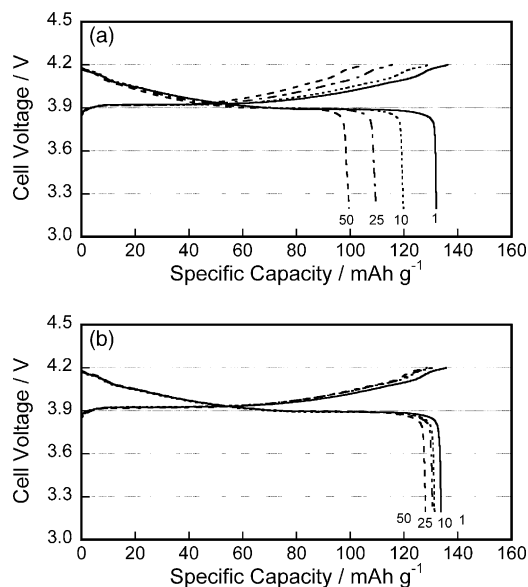

Fig. 7. Charge–discharge curves of the Li/Li[TFSI] cells with an ionic liquid/Li at 298 K: (a) EMI[FSI]; (b) Py$_{13}$[FSI]. [Li[TFSI]] = 0.3 mol kg^{-1} (3.2–4.2 V, constant current mode). Cycle number is indicated in the figure. Charge–discharge current rate 0.1 C.

3.4. Cycling properties over 50 cycles of the Li/LiCoO$_2$ cell

Fig. 7 shows the cycle dependencies of the discharge capacities of Li/LiCoO$_2$ cells at the 0.1 C rate. In the EMI[FSI] case, the discharge capacitance falls as the cycles continue, even though, as stated above, the addition of Li[TFSI] apparently improved the electrochemical stability. On the other hand, a much more stable cycling over 50 cycles occurred when the Py$_{13}$[FSI] electrolyte was used instead of the EMI[FSI]. This result suggests that the electrochemical stability of the starting neat RTILs can influence the cell performance.

4. Conclusion

An investigation of the physical properties of RTILs based on $[FSI]^-$, with and without 0.3 mol kg^{-1} of Li[TFSI], and a charge–discharge test of a Li/LiCoO$_2$ cell containing the $[FSI]^-$ melts produced many results which were unique when compared to those obtained for $[TFSI]^-$ melts:

(1) The viscosity of the neat $[FSI]^-$ melts was about 60% of that of the corresponding $[TFSI]^-$ melts. It must be noted that the undesired increase in the viscosity of the RTILs with the addition of a lithium salt was about 50% suppressed in the $[FSI]^-$ melts compared to those of the corresponding $[TFSI]^-$ melts.
(2) With the use of the low-viscosity $[FSI]^-$ melts, the rate operation of a Li/LiCoO$_2$ cell containing RTILs only was greater than 1 C.
(3) Since the best C–D property was observed in the EMI[FSI] sample, which exhibited the poorest electrochemical stability of all the $[FSI]^-$ melts tested in this study, good SEI films must have been formed on the electrode materials.

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