

# Accepted Manuscript

Charge-discharge Characteristics of Natural Graphite Electrode in *N, N*-Diethyl- *N*-Methyl-*N*-(2-methoxyethyl)ammonium Bis(trifluoromethylsulfonyl)amide Containing Lithium Ion for Lithium-ion Secondary Batteries

Jun Towada, Tomohiro Karouji, Hiroki Sato, Yoshihiro Kadoma, Kazuaki Shimada, Koichi Ui

PII: S0378-7753(14)01710-8

DOI: [10.1016/j.jpowsour.2014.10.101](https://doi.org/10.1016/j.jpowsour.2014.10.101)

Reference: POWER 20021

To appear in: *Journal of Power Sources*

Received Date: 25 July 2014

Revised Date: 7 October 2014

Accepted Date: 16 October 2014

Please cite this article as: J. Towada, T. Karouji, H. Sato, Y. Kadoma, K. Shimada, K. Ui, Charge-discharge Characteristics of Natural Graphite Electrode in *N, N*-Diethyl- *N*-Methyl-*N*-(2-methoxyethyl)ammonium Bis(trifluoromethylsulfonyl)amide Containing Lithium Ion for Lithium-ion Secondary Batteries, *Journal of Power Sources* (2014), doi: 10.1016/j.jpowsour.2014.10.101.

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.





**Charge-discharge Characteristics of Natural Graphite Electrode in *N, N*-Diethyl-*N*-Methyl-*N*-(2-methoxyethyl)ammonium Bis(trifluoromethylsulfonyl)amide Containing Lithium Ion for Lithium-ion Secondary Batteries**

Jun TOWADA <sup>a</sup>, Tomohiro KAROUJI <sup>b</sup>, Hiroki SATO <sup>b</sup>,

Yoshihiro KADOMA <sup>b</sup>, Kazuaki SHIMADA <sup>c</sup>, and Koichi UI <sup>b,\*</sup>

<sup>a</sup> *Department of Frontier Matter and Function Engineering, Graduate School of Engineering, Iwate University, Morioka, Iwate 020-8551, Japan.*

<sup>b</sup> *Department of Frontier Materials and Function Engineering, Graduate School of Engineering, Iwate University, Morioka, Iwate 020-8551, Japan.*

<sup>c</sup> *Department of Chemistry and Bioengineering, Graduate School of Engineering, Iwate University, 4-3-5 Ueda, Morioka, Iwate 020-8551, Japan.*

- **Article type** **Short Communication**
- **\*Corresponding author** Associate Professor Koichi UI (M)
- **Business Affiliation** Iwate University
- **Full postal address** 4-3-5 Ueda, Morioka, Iwate 020-8551, Japan

- Phone and fax number +81-19-621-6340 / 6314
- E-mail address kui@iwate-u.ac.jp

ACCEPTED MANUSCRIPT

### Abstract

We investigated the electrochemical characteristics of a natural graphite electrode in room-temperature ionic liquids not containing additives. *N,N*-diethyl-*N*-methyl-*N*-(2-methoxyethyl)ammonium bis(trifluoromethylsulfonyl)amide (DEME-TFSA) containing lithium bis(trifluoromethylsulfonyl)amide (Li-TFSA) as the electrolyte and a natural graphite electrode as the negative electrode material were employed. The charge-discharge tests showed that the discharge capacity and the charge-discharge efficiency of the natural graphite electrode at the 1st cycle were 318 mAh g<sup>-1</sup> and 75.6%, respectively. The cycle performance showed that the discharge capacity and the charge-discharge efficiency were stably maintained at *ca.* 320 mAh g<sup>-1</sup> and 100%, respectively, until the initial 10th cycle. The *ex-situ* X-ray diffraction measurements showed that lithium-graphite intercalation compounds, such as LiC<sub>12</sub> and LiC<sub>6</sub>, were formed after the 1st charge. The structural change in the natural graphite electrode was reversible because graphite recovered to its original structure after the 1st discharge. These results clarified that the graphite electrode could operate as a negative electrode for lithium-ion secondary batteries in DEME-TFSA containing Li-TFSA without organic solvents.

**Keywords:** Lithium-ion secondary battery, Room-temperature ionic liquid, Bis(trifluoromethylsulfonyl)amide, Negative electrode, Natural graphite

**Highlights:**

- Initial characteristics of graphite at 25°C with a capacity close to theoretical.
- A stable cycle performance was obtained.
- DEME-TFSA containing Li-TFSA provides reversible lithium ion intercalation/extraction without any additives.
- DEME-TFSA containing Li-TFSA may provide suitable SEI film.
- DEME-TFSA containing Li-TFSA may prevent undesirable side reactions.

## 1. Introduction

In recent years, the application of lithium-ion secondary batteries is expected as an automotive power supply and as an electric energy storage system for grid-connection with new energy resources. Further improvement in its safety is necessary because there were ignition accidents involving lithium-ion secondary batteries. Thus, novel electrolytes and electrode materials are being developed. In order to enhance the thermal stability of the electrolyte, ambient-temperature molten salts (room-temperature ionic liquids, RTILs) having the advantages of a low volatility and nonflammability have been extensively studied as one solution.

However, graphite, which has been used as a major negative electrode material, does not sufficiently operate in many types of RTILs. The main reason would be due to the electrochemical organic cation intercalation into the graphite layers [1, 2] before the formation of an effective solid electrolyte interface (SEI) film on the graphite particles during the 1st charging. Recently, it has been reported that the graphite electrode indicated good charge-discharge characteristics by adding an additive, such as vinylene carbonate [3], to the RTILs or using the RTILs, such as 1-ethyl-3-methylimidazolium bis(fluorosulfonyl)amide (EMI-FSA) [4] and *N*-methyl-*N*-propylpyrrolidinium bis(fluorosulfonyl)amide (P<sub>13</sub>-FSA) [5].

On the other hand, there is *N, N*-diethyl-*N*-methyl-*N*-(2-methoxyethyl)ammonium bis(trifluoromethylsulfonyl)amide (DEME-TFSA) among the TFSA-based RTILs. It has been reported that the charge-discharge characteristics of LiCoO<sub>2</sub> showed values near the theoretical charge-discharge capacity in the first cycle and an excellent reversibility for the lithium bis(trifluoromethylsulfonyl)amide (Li-TFSA) / DEME-TFSA electrolyte [6]; the graphite electrode achieved a reasonable cycle performance by adding vinylene carbonate as an additive to these RTIL electrolytes [7]. Nevertheless, the present state is that the electrochemical characteristics of a graphite electrode in the TFSA-based RTILs without organic solvent additives are poor [5, 8]. It is known that the thermal stability of TFSA-based RTILs is higher than that of FSA-based RTILs [9].

In order to utilize the features of the RTILs, we employed TFSA-based RTILs. In this study, we investigated DEME-TFSA containing Li-TFSA as an electrolyte. In order to obtain the basic knowledge needed for the design of lithium-ion secondary batteries using RTILs as the electrolyte, the electrochemical characteristics (charge-discharge characteristics) of the natural graphite negative electrode in DEME-TFSA containing Li-TFSA were evaluated.

## 2. Experimental

The graphite electrode was prepared in a way similar to that previously reported [10, 11]. Natural graphite powder (Kansai Coke and Chemical Co., 3  $\mu\text{m}$  average diameter, NG-3) as the active material and poly (vinylidene fluoride) (Kureha. Co., KF polymer #9130, PVdF) as the binder were employed. A suspension composed of 90 wt. % NG-3 powder and 10 wt. % PVdF was prepared using *N*-methyl-2-pyrrolidone (Kanto Chemical Co., degree of purity 99.0%, NMP) as the dispersing medium. It was then stirred in a reagent bottle at room temperature for 24h. It was next coated on a Cu foil (Housen, thickness 18  $\mu\text{m}$ ) using an automatic coating machine (Housen, MC20). The electrode was then pressed using a small-sized press machine (AS ONE, AH-2003, 10min, 10t, room temperature) after an initial air-drying at 80°C for 1h. The NG-3 electrode was then punched into a circular shape and dried under vacuum at 200°C for 3h.

An R2032 coin-type cell, which consisted of the NG-3 electrode (99.93 mm<sup>2</sup>, W.E.), a pressed lithium metal foil (C.E.), 1 mol dm<sup>-3</sup> (0.7 mol kg<sup>-1</sup>) Li-TFSA (Morita Chemical Industries Co., 99%) / DEME-TFSA (Kanto Chemical Co., 99%) as the electrolyte, and Celgard#2500 as the separator was assembled in a glove box (Miwa Mtg Co., DBO-1NKP-1U-2, dew point -80°C) filled with dry argon at 25°C.

Cyclic voltammetry was performed using a computer-controlled electrochemical

measuring system (Hokuto Denko, HZ-5000). The cyclic voltammograms were measured at the scan rate of  $0.1 \text{ mV s}^{-1}$  between the voltages of 5 mV and 2.0 V at  $25^\circ\text{C}$ . The charge-discharge cycle tests were performed using an automatic battery charging-discharging instrument (Hokuto Denko, HJR-1010mSM8) at the current density of  $35 \text{ mA g}^{-1}$  ( $1C = 350 \text{ mA g}^{-1}$ ) between 5 mV and 2.0 V at  $25^\circ\text{C}$ .

The crystal structure of the samples was examined by X-ray diffraction (XRD) using an X-ray diffractometer (Bruker, D8 ADVANCE, radiation source Cu  $K\alpha$ , ( $\lambda = 0.15418 \text{ nm}$ ), tube voltage 40 kV). The electrodes were charged using the above automatic battery charging-discharging instrument to obtain the samples for identification. The charge condition was controlled as follows: the charge voltage was maintained for 12h in the constant voltage (CV) mode after it reached 5 mV in the constant current (CC) mode. It was discharged to 2.0 V in the CC mode after CC-CV mode charging. The discharge process involves the deintercalation reaction of Li(I) ions which recovers the structure of graphite. The CC mode was thus employed because the voltage did not need to remain constant. The obtained samples were washed with diethyl carbonate (Wako Chemical Industries, Ltd., 98.0+% (Capillary GC), DEC) if the samples needed to be washed and then dried for 12h under reduced pressure. The samples were examined in an airtight sample holder to prevent any contact of the

sample with air.

The surface analysis of the NG-3 electrodes was carried out by X-ray photoelectron spectroscopy (XPS) analysis (Perkin-Elmer, PHI 5600,  $\phi = 800 \mu\text{m}$ ) with Al  $K\alpha$  radiation ( $h\nu = 1486 \text{ eV}$ ). The samples were prepared under the same conditions for the XRD analysis. They were put in a transfer vessel to prevent completely contact of the samples with air until the samples were introduced into XPS.

### 3. Results and discussion

Fig. 2 shows the cyclic voltammograms of the NG-3 electrode ( $1.23 \text{ mg cm}^{-2}$ ) in  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA at  $25^\circ\text{C}$ . The irreversible reduction peak at the 1st cycle is observed around 1.5 V. In addition, the reduction current at voltages from 1.5 to 0.2 V for the 1st cycle is greater than that after the 2nd cycle. The  $\text{LiC}_x$  phase is formed below 0.2 V [12], indicating that the above reduction current would be due to the decomposition of the electrolyte. Fig. 3 shows the high-resolution XPS spectra of the  $\text{F}_{1s}$  region for the NG-3 electrodes (a) before immersion and (b) after charging to 1.5 V. As shown in Fig. 3(a), the peak was detected at around 688 eV on the surface of the NG-3 electrodes. The peak at around 688 eV was assigned to the binding energy of a C-F\* bond derived from the PVdF binder [13]. As shown in Fig. 7(b), the peaks were

detected at around 685.5 eV and 688.5 eV. The peak at around 685.5 eV was assigned to a Li-F\* bond [14, 15]. The peak at around 688.5 eV was assigned to a C-F\* bond derived from -CF<sub>3</sub>\* (TFSA<sup>-</sup> anion) [14]. It is considered that LiF would be formed due to the reductive decomposition of TFSA<sup>-</sup> anion because the peak of Li-F\* bond was detected at around 685.5 eV. In the case of several RTILs, several researches on the electrochemical reduction of the polyatomic anion such as AlCl<sub>4</sub><sup>-</sup> [1, 10, 11] and TFSA<sup>-</sup> [14-16] have been reported. These reports support the above consideration. In contrast, the reductive decomposition of DEME cation is not yet judged. Based on these results, it was suggested that the deposit would be formed on the NG-3 electrode due to the reductive decomposition of TFSA<sup>-</sup> anion at around 1.5 V at least. The reduction and oxidation waves are observed in the voltage ranges from approximately 0.2 to 0.0 V and from 0.2 to 0.4 V, respectively. It is considered that the above peaks correspond to the intercalation and deintercalation reactions of the Li(I) ions into and from the graphite layer [12], respectively. After the initial 2 cycles, the redox wave shapes are almost similar. The irreversible reduction wave around 0.2 to 1.5 V considered to be due to the reduction of the electrolyte is not observed. Based on these results, it is suggested that the intercalation and deintercalation reactions of the Li(I) ions into and from the NG-3 electrodes in 1 mol dm<sup>-3</sup> Li-TFSA / DEME-TFSA would occur.

Fig. 4 shows the charge-discharge curves of the NG-3 electrodes ( $1.20 \text{ mg cm}^{-2}$ ) at  $25^\circ\text{C}$  at the constant current density of  $35 \text{ mA g}^{-1}$  ( $0.1C$ ) in  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA. In this paper, the cathodic polarization due to the Li(I) ion intercalation into the graphite layer is defined as the charge and anodic polarization due to the Li(I) ion deintercalation is defined as the discharge. The discharge capacity and the charge-discharge efficiency at the 1st cycle were  $318 \text{ mAh g}^{-1}$  and 75.6%, respectively. These values are almost similar to those in the typical organic solvent electrolytes. It is observed that the shape of the charge curve for the 1st cycle is different from that of the charge curves after the 2nd cycle in the voltage range from 0.2 to 1.5 V. A similar behavior is observed in the CV (Fig. 2). This behavior has also been confirmed in organic solution electrolytes [18, 19] and the chloroaluminate type RTIL electrolyte [10, 11]. Thus, it is considered that the reduction reaction of the electrolyte could be suppressed after the 2nd cycle because an interface similar to the SEI film on the surface of the graphite particles would be formed during the 1st cycle. Thus, it is considered that the voltage shoulder on the 1st charge-discharge curve would indicate a side reaction due to the reduction of the electrolyte. Based on these results, it was found that the NG-3 electrode operated as a negative electrode in  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA for lithium-ion secondary batteries.

Fig. 5 shows the cycle performance of the NG-3 electrodes at 25°C at a constant current density of 35 mA g<sup>-1</sup> (0.1C). The discharge capacity maintained *ca.* 320 mAh g<sup>-1</sup> during the initial 10 cycles. It is interesting to note that the charge-discharge efficiency significantly increased from the 1st cycle to the 2nd cycle and then maintained *ca.* 100% after the 3rd cycle. This suggests that the reduction of the electrolyte would almost not occur as a side reaction after the 2nd cycle.

Fig. 6 shows the *ex-situ* XRD patterns of the NG-3 electrode during the 1st charge-discharge. As shown in Fig. 6(i), in the case of the NG-3 electrode soaked in the electrolyte, washed, and then dried, the (002) and (004) peaks were observed at 26.4° (*d* = 338 pm) and 54.6° (*d* = 168 pm), respectively, were ascribed to the diffraction peaks of the natural graphite. As shown in Fig. 6(ii), the diffraction peaks of the natural graphite shifted to the lower positions of 24.1° (*d* = 370 pm) and 49.3° (*d* = 185 pm) corresponding to (001) and (002), respectively, after charging to 5 mV. This indicates that the distance between the graphite layers increased due to the Li(I) ion intercalation after charging and the stage 1 intercalation compound, LiC<sub>6</sub>, is formed. These values correspond to the literature values [10, 20, 21]. In addition, a diffraction peak is appeared at 25.4°, suggesting that the stage 2 intercalation compound, LiC<sub>12</sub>, is formed [10, 20]. Thus, it is considered that the stage 1 and stage 2 coexist. As shown in Fig.

6(iii), the diffraction peaks shifted to the higher positions of  $26.4^\circ$  ( $d = 338$  pm) and  $54.6^\circ$  ( $d = 168$  pm) corresponding to (002) and (004), respectively, after discharging to 2.0 V. Based on these results, it was clarified that the structural change in the NG-3 electrode due to the intercalation and deintercalation of Li(I) ions occurred in a reversible manner and a lithium-graphite intercalation compound was formed. Considering the 1st charge-discharge efficiency of 75.6%, the formation of a side reaction would be considered. It seems reasonable to postulate that an SEI film is formed because a Li-F\* bond was detected by XPS. However, in the *ex-situ* XRD patterns of this study, no other crystalline product was detected. Concerning the formation of inorganic compounds on the graphite surface from the RTILs, several results have been previously reported. In the chloroaluminate type RTIL electrolyte, the formation of LiCl on the graphite surface was observed during the 1st charge [10, 11]. Sugimoto *et al.* reported that the inorganic compound layer generated from the EMI-FSA system was thinner than that derived from a typical organic solvent electrolyte as a result of the XPS analysis [17]. It is thus necessary to identify in more detail the surface product mainly focusing on an organic component using other analytical methods.

Fig. 7 shows the photographs of the NG-3 electrodes (a) after immersion and (b)

after discharging to 5 mV. The surface of the NG-3 electrode was a black color before charging. In contrast, the surface of the NG-3 electrode after charging to 5 mV changed to a golden yellow color. This result demonstrates that the 1st stage intercalation compound,  $\text{LiC}_6$ , would be formed according to the electrochemical intercalation of the  $\text{Li(I)}$  ion into the graphite layer [12].

#### 4. Conclusion

In order to obtain the basic knowledge needed for the design of lithium-ion secondary batteries using the RTILs as the electrolyte, the electrochemical characteristics of the natural graphite negative electrode in DEME-TFSA containing Li-TFSA was evaluated. The initial charge-discharge characteristics and the cycle performance of the NG-3 electrode in the  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA electrolyte were almost similar to those in a typical organic solvent electrolyte. The structural changes between graphite and a lithium-graphite intercalation compound during the charge-discharge reaction showed a reversible reaction although a side reaction occurred during the 1st charge. This indicated that the electrochemical intercalation and deintercalation reactions of the lithium ions into and from the graphite layer in DEME-TFSA containing lithium ions occur in a reversible manner.

These results lead to the conclusion that the graphite electrode can operate as a negative electrode for lithium-ion secondary batteries using RTILs consisting of the TFSA<sup>-</sup> anion without organic solvents. In conclusion, it is important to clarify the electrochemical behavior between the RTILs consisting of the TFSA<sup>-</sup> anion and the graphite electrode because DEME-TFSA containing Li-TFSA may provide suitable SEI film.

#### **Acknowledgments**

This study was partially supported by a Grant-in-Aid for Scientific Research (C) from the Japanese Ministry of Education, Science, Sports and Culture (No. 25410248) and A-step (Exploratory Research) of Japan Science and Technology Agency (JST).

#### **References**

- 1) R. T. Carlin, J. Fuller, W. K. Kuhn, M. J. Lysaght, and P. C. Trulove, *J. Appl. Electrochem.*, 26 (1996) 1147.
- 2) T. E. Sutto, P. C. Trulove, and H. C. DeLong, *Electrochem. Solid-State Lett.*, 6 (2003) A50.
- 3) H. Zheng, K. Jiang, T. Abe, and Z. Ogumi, *Carbon*, 44 (2006) 203.

- 4) M. Ishikawa, T. Sugimoto, M. Kikuta, E. Ishiko, and M. Kono, *J. Power Sources*, 162 (2006) 658.
- 5) S. Seki, Y. Kobayashi, H. Miyashiro, Y. Ohno, Y. Mita, N. Terada, P. Charest, A. Guerfi, and K. Zaghib, *J. Phys. Chem. C*, 112 (2008) 16708.
- 6) S. Seki, Y. Kobayashi, H. Miyashiro, Y. Ohno, Y. Mita, A. Usami, N. Terada, and M. Watanabe, *Electrochem. Solid-State Lett.*, 8 (2005) A577.
- 7) T. Sato, T. Maruo, S. Marukane, and K. Takagi, *J. Power Sources*, 138 (2004) 253.
- 8) M. Egashira, A. Kanetomo, N. Yoshimoto, and M. Morita, *Electrochemistry*, 78 (2010) 370.
- 9) M. Ishikawa, T. Sugimoto, M. Kikuta, E. Ishiko, and M. Kono, *J. Power Sources*, 162 (2006) 658.
- 10) K. Ui, J. Towada, S. Agatsuma, N. Kumagai, K. Yamamoto, H. Haruyama, K. Takeuchi, and N. Koura, *J. Power Sources*, 196 (2011) 3900.
- 11) J. Towada, Y. Kadoma, K. Shimada, N. Kumagai, and K. Ui, *Electrochemistry*, 81 (2013) 432.
- 12) M. Letellier, F. Chevallier, and M. Morerette, *Carbon*, 45 (2007) 1025.
- 13) D. Aurbach, B. Markovsky, A. Shechter, Y. Ein-Eli, and H. Cohen, *J. Electrochem. Soc.*, 143, 3809 (1996).
- 14) P. C. Howlett, N. Brack, A. F. Hollenkamp, M. Forsyth, and D. R. MacFarlane, *J.*

- Electrochem. Soc., 153, A595 (2006).
- 15) L. Zhao, J. Yamaki, and M. Egashira, *J. Power Sources*, 174, 352 (2007).
  - 16) C. C. Nguyen and S.-W. Song, *Electrochem. Commun.*, 12, 1593 (2010).
  - 17) T. Sugimoto, M. Kikuta, E. Ishiko, M. Kono, and M. Ishikawa, *J. Power Sources*, 183 (2008) 436.
  - 18) K. Ui, D. Fujii, Y. Niwata, T. Karouji, Y. Shibata, Y. Kadoma, K. Shimada, and N. Kumagai, *J. Power Sources*, 247 (2014) 981.
  - 19) J. Drogenik, M. Gaberscek, R. Dominko, F. W. Poulsen, M. Mongensen, S. Pejovnik, and J. Jamnik, *Electrochim. Acta*, 48 (2003) 883.
  - 20) K. Ui, S. Kikuchi, F. Mikami, Y. Kadoma, and N. Kumagai, *J. Power Sources*, 173 (2007) 518.

**All figure captions**

**Fig. 1.** Structural formula of DEME-TFSA.

**Fig. 2.** Cyclic voltammograms of the NG-3 electrode using PVdF 10 wt. % as the binder in the  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA electrolyte at  $25^\circ\text{C}$ ; voltage range: 5 mV to 2.0 V, scan rate:  $0.1 \text{ mV s}^{-1}$ ; (—) 1st cycle; (---) 2nd cycle; (.....) 3rd cycle.

**Fig. 3.** High-resolution XPS spectra (solid line) and their fitting curves (dashed line) of the  $F_{1s}$  region for the NG-3 electrode using PVdF 10 wt. % as the binder in the  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA electrolyte; (a) before immersion and (b) after charging to 1.5 V; (---)  $\text{LiF}^*$ ; (- · -)  $\text{C-F}^*$  (PVdF); (- · · -)  $\text{CF}_3^*$  (TFSA).

**Fig. 4.** Initial charge-discharge curves of the NG-3 electrode using PVdF 10 wt. % as the binder in the  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA electrolyte at  $25^\circ\text{C}$ ; voltage range: 5 mV to 2.0 V, current density:  $35 \text{ mA g}^{-1}$  (0.1C); (—) 1st cycle; (---) 2nd cycle; (.....) 3rd cycle.

**Fig. 5.** Cycle performance of the NG-3 electrode using PVdF 10 wt. % as the binder in the  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA electrolyte at  $25^\circ\text{C}$ ; voltage range: 5 mV to 2.0 V, current density:  $35 \text{ mA g}^{-1}$  (0.1C); (○) discharge capacity; (●) charge-discharge efficiency.

**Fig. 6.** *Ex-situ* XRD patterns of the NG-3 electrode using PVdF 10 wt. % as the binder in the  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA electrolyte; (i) after immersion, (ii) after charging to 5 mV, and (iii) after discharging to 2.0 V.

**Fig. 7.** Photographs of the NG-3 electrode using PVdF 10 wt. % as the binder in the  $1 \text{ mol dm}^{-3}$  Li-TFSA / DEME-TFSA electrolyte; (a) after immersion, (b) after charging to 5 mV.

## Figures

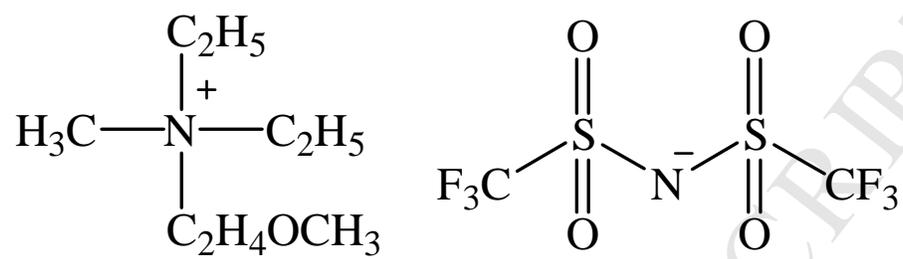
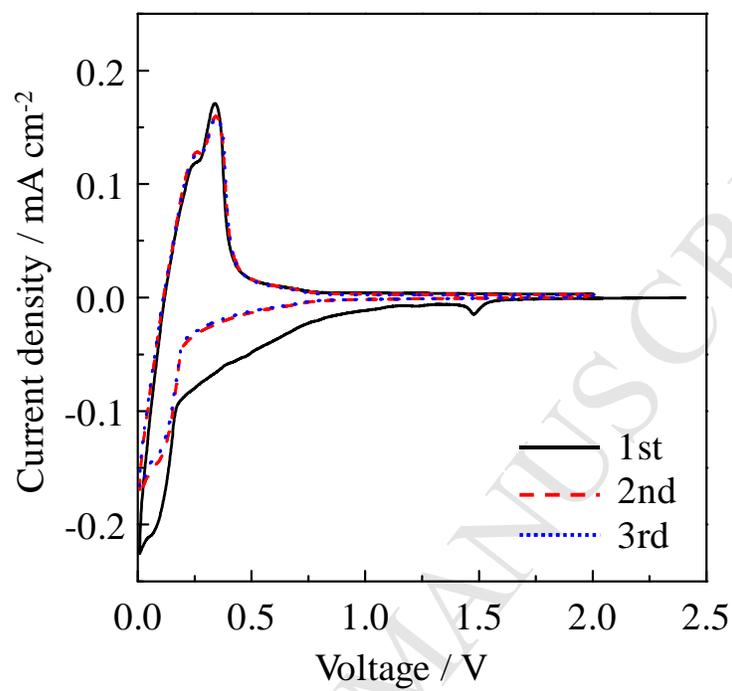


Fig. 1.

**Fig. 2.**

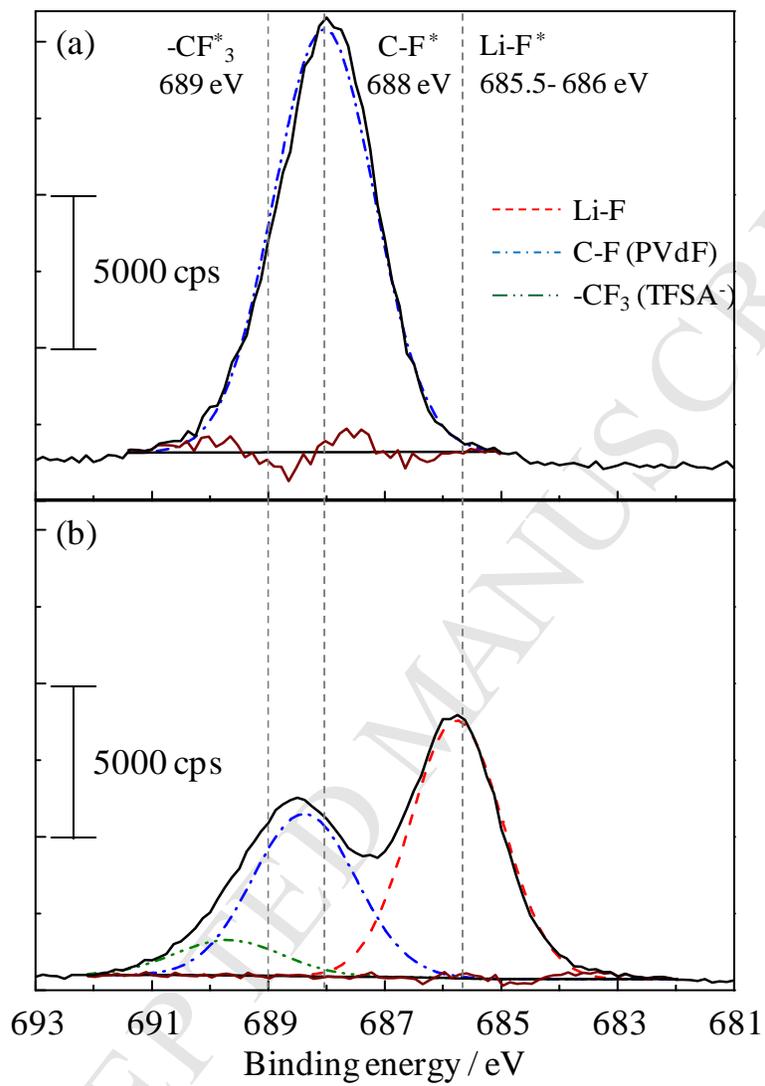
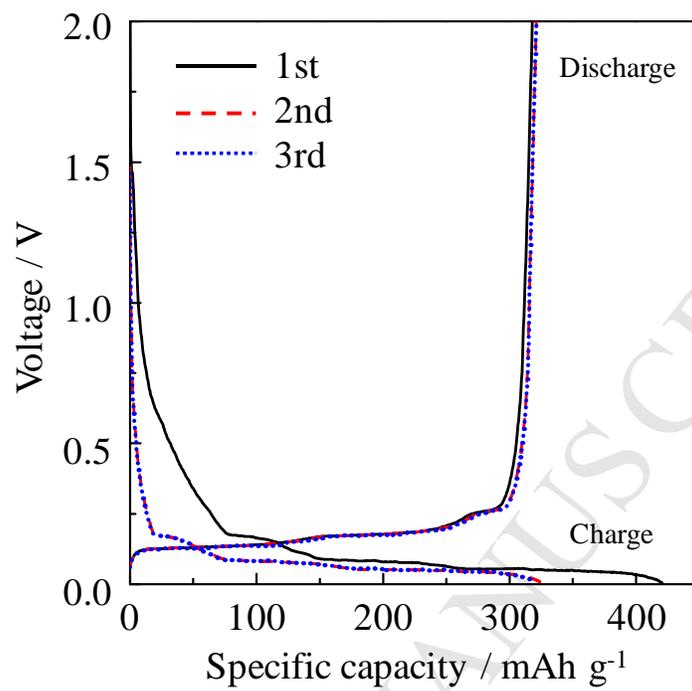


Fig. 3.

**Fig. 4.**

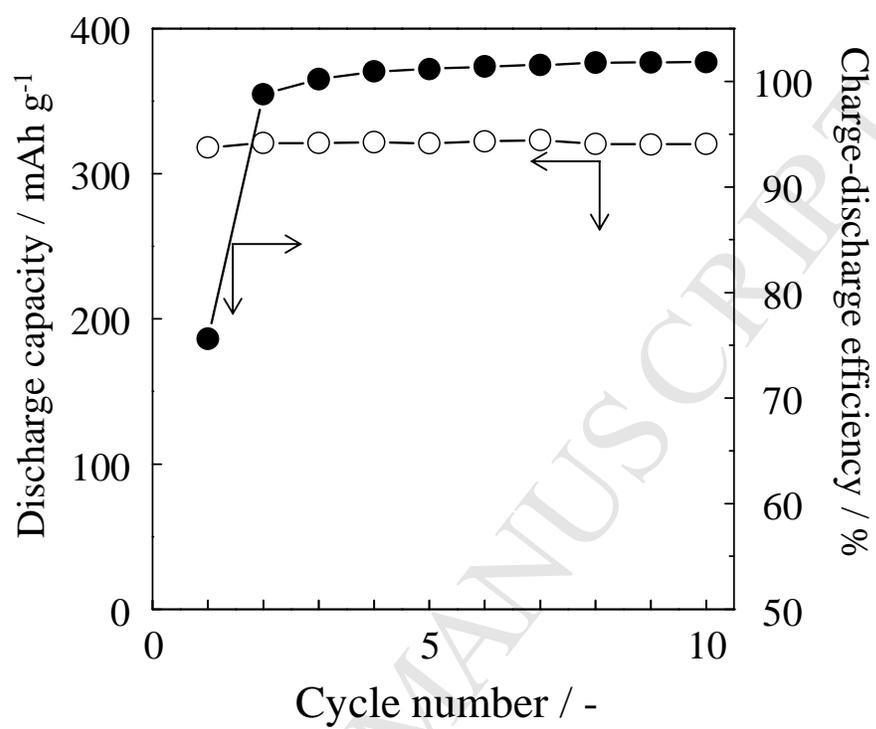


Fig. 5.

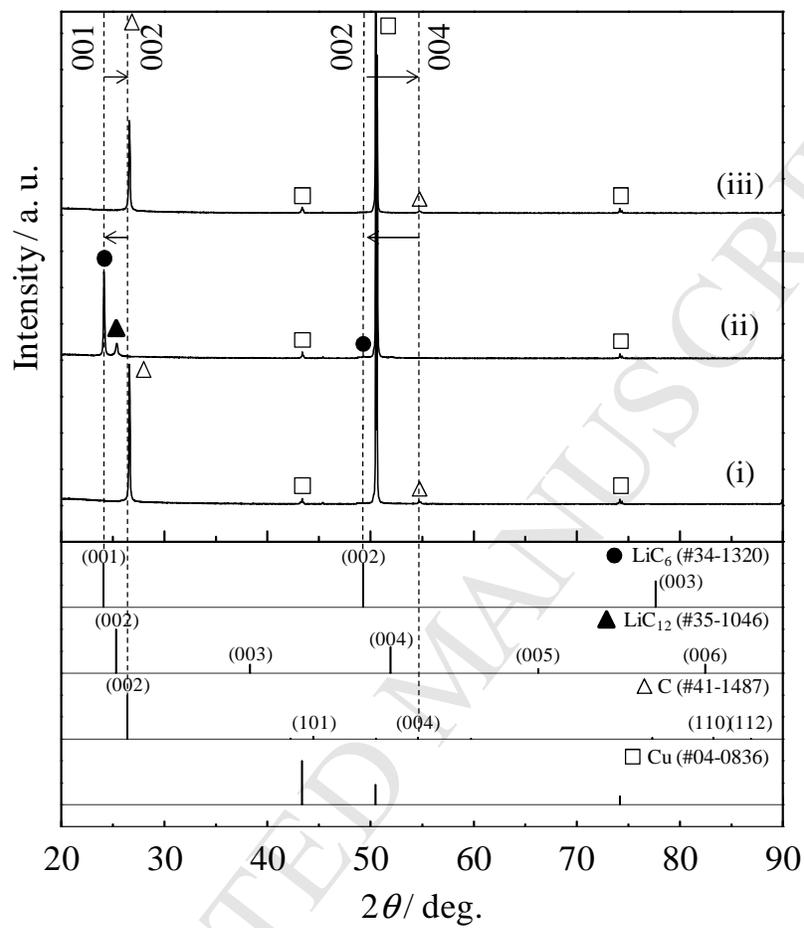
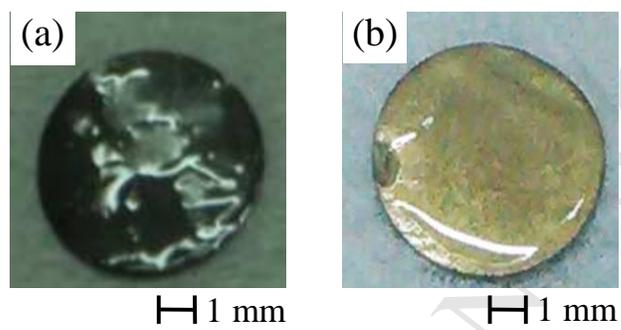


Fig. 6.



**Fig. 7.**

**Highlights:**

- Initial characteristics of graphite at 25°C with a capacity close to theoretical.
- A stable cycle performance was obtained.
- DEME-TFSA containing Li-TFSA provides reversible lithium ion intercalation/extraction without any additives.
- DEME-TFSA containing Li-TFSA may provide suitable SEI film.
- DEME-TFSA containing Li-TFSA may prevent undesirable side reactions.