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Short communication

# Cycling performance of lithium metal polymer cells assembled with ionic liquid and poly(3-methyl thiophene)/carbon nanotube composite cathode

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## Abstract

A poly(3-methylthiophene) (PMT)/multi-walled carbon nanotube (CNT) composite is synthesized by *in situ* chemical polymerization. The PMT/CNT composite is used as an active cathode material in lithium metal polymer cells assembled with ionic liquid (IL) electrolytes. The IL electrolyte consists of 1-ethyl-3-methylimidazolium tetrafluoroborate ($EMIBF_4$) and $LiBF_4$. A small amount of vinylene carbonate is added to the IL electrolyte to prevent the reductive decomposition of the imidazolium cation in $EMIBF_4$. A porous poly(vinylidene fluoride-*co*-hexafluoropropylene) (P(VdF-*co*-HFP)) film is used as a polymer membrane for assembling the cells. Electrochemical properties of the PMT/CNT composite electrode in the IL electrolyte are evaluated and the effect of vinylene carbonate on the cycling performance of the lithium metal polymer cells is investigated. The cells assembled with a non-flammable IL electrolyte and a PMT/CNT composite cathode are promising candidates for high-voltage–power sources with enhanced safety.

*Keywords:* Carbon nanotube; Conducting polymer; Ionic liquid; Lithium metal polymer cell; Poly(3-methylthiophene); Safety

## 1. Introduction

Rechargeable lithium batteries that employ lithium metal as a negative electrode (anode) are considered to be one of the most important prospects for next-generation power sources for portable electronic devices and electric vehicles [1]. This is because lithium metal offers the largest specific capacity and specific energy as a negative electrode among known materials. Thus, many research efforts have been devoted to the development of lithium metal batteries. In particular, serious safety issues surrounding such batteries must be solved before they are utilized widely, especially in large applications such as electric vehicles.

The highly flammable organic solvents commonly employed in lithium batteries are recognized as one of the main causes of serious safety drawbacks [2,3]. In the quest for a non-flammable electrolyte system, ionic liquids (ILs) have been recognized as one of the safest electrolytes that can be used in rechargeable lithium batteries [3,4]. ILs are often liquid at room and sub-ambient temperatures. They have negligible vapour pressure, wide electrochemical stability, non-flammability, high-ionic conductivity, and high-thermal stability. Hence, these ILs are highly attractive as an electrolyte material for safe lithium batteries. Various ILs have been synthesized and tested for application in lithium batteries [5–22]. Among them, a family of imidazolium salts has been widely studied because of their low

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viscosity, high solubility of lithium salt, high-ionic conductivity and high-anodic stability. On the other hand, these imidazolium-based ILs have low stability at the positive electrode (cathode) and hence a small quantity of film-forming additive has to be introduced into the electrolyte to prevent its cathodic decomposition [8,11,17,22].

Inorganic lithium intercalation materials such as $LiCoO_2$, $LiNiO_2$ and $LiMn_2O_4$ for cathodes in lithium batteries have been the subject of intense research activity for more than a decade [1,23,24]. Conducting polymers such as polyacetylene, polypyrrole and polyaniline also have the potential to be applied as cathode materials and offer the prospect of low cost and simple manufacturing processes [25]. These conducting polymers, however, have been much less studied because of conductivity limitations in their reduced state. In recent work, we have begun to investigate polyaniline (PANI)/multi-walled carbon nanotube (CNT) composite materials in which the CNT matrix provides a percolated conduction pathway for the conducting polymer [22,26]. The latter is polymerized on to the carbon nanotubes as a ~25 nm thick coating such that, even in the reduced state, the overall conduction pathway is not highly resistive and substantial rates become possible in cells using this as a cathode active material. The remaining limitation in these materials was the low-potential range available from the composite cathode. Most conducting polymer-based cathodes have low-redox potentials, and hence are unable to provide a battery capable of delivering a high voltage that is comparable with inorganic cathode materials. Among the possible conducting polymers that can be used as cathode materials, poly(3-methylthiophene) (PMT) is of particular interest because it is known to be stable in its doped and undoped states and its redox potential is higher than 3.6 V vs. Li [27]. Hence, in the work presented here, an investigation has been made of the electrochemical performance of PMT as a cathode material for a lithium metal cell using an IL electrolyte. The PMT/CNT composite is synthesized by a chemical method. To the best of our knowledge, there has been no report on the use of a PMT/CNT cathode and an IL electrolyte for lithium metal batteries.

## 2. Experimental

### 2.1. Preparation of PMT/CNT composite material

Multi-walled CNT was purchased from ILJIN Nanotech. It had been prepared by chemical vapour deposition and its aspect ratio was about 1000. A PMT/CNT nanocomposite was produced via *in situ* chemical synthesis. In a typical experiment, 2.0 g of CNT was suspended in 800 ml of acetonitrile containing 19.2 ml of 3-methylthiophene (0.25 M). The suspension was sonicated for 3 h in an ice bath, to facilitate good dispersion of CNT. After sonication, the CNT-suspended monomer solution was transferred to the polymerization reactor, which was kept at 0 °C. To this suspension, about 800 ml of acetonitrile containing 64.9 g of $FeCl_3$ (0.5 M) was added drop-by-drop for 3 h, with mechanical stirring at 300 rpm. After the addition was completed, the stirring was continued overnight at the same bath temperature (0 °C). The resulting PMT/CNT composite precipitate was filtered, washed successively with acetonitrile, water and methanol, and finally dried at 70 °C for 12 h.

### 2.2. Cell assembly

To prepare the cathode, the PMT/CNT composite material (85 wt.%) was initially mixed with Super-P carbon (MMM Co., 8 wt.%), and then the dry mixture was ball-milled at 150 rpm for 12 h. After the dry ball-milling, a binder solution containing poly(vinylidene fluoride) (PVdF, 7 wt.%) dissolved in *N*-methyl-2-pyrrolidone (NMP) was mixed with the dry mixture, and the milling was continued for a further 12 h. The resultant viscous slurry was cast on Al foil to a thickness of 60 μm using a doctor blade. The electrode was air dried overnight and then dried under vacuum at 70 °C for 12 h. Finally, the electrodes were roll pressed to enhance particulate contact and adhesion to the foil. The roll-pressed electrodes had a polymer coating thickness of ~25 μm. The anode consisted of a lithium foil of 100 μm thick (Honjo Metal Co. Ltd.) that was pressed on to a copper current-collector.

A porous poly(vinylidene fluoride-*co*-hexafluoropropylene) (P(VdF-*co*-HFP)) membrane was prepared as follows. P(VdF-*co*-HFP) (Kynar 2801), fumed silica, dibutyl phathalate (DBP) and acetone were mixed together and ball-milled for 48 h and then cast to a thickness of 500 μm using a doctor blade. After 30 min, the membranes were immersed in methanol to remove the DBP. The membranes were then vacuum dried at 60 °C for 12 h. Before using the P(VdF-*co*-HFP) membrane in the cell assembly, it was immersed in the IL-based electrolyte for 5 min. The IL electrolyte was prepared by dissolving 1.0 M $LiBF_4$ in 1-ethyl-3-methylimidazolium tetrafluoroborate ($EMIBF_4$). Vinylene carbonate was added to the IL electrolyte at a concentration of 10 wt.% to serve as a film-forming agent. Lithium metal polymer cells were assembled by sandwiching the P(VdF-*co*-HFP) membrane soaked in the IL electrolyte between a lithium anode and a PMT/CNT composite cathode. The cell assembly was carried out in a dry box filled with argon gas. After assembly, the cells were kept at 60 °C for 12 h to allow the PMT/CNT composite cathode to become imbued with the IL electrolyte.

### 2.3. Measurements

The morphologies of pure CNT and the PMT/CNT composite were examined by means of a scanning electron microscope (PHILIPS, XL30SFEG). Linear sweep voltammetry was performed to investigate the electrochemical stability window of the IL-based electrolyte on a stainless-steel (SS) working electrode at a scanning rate of 1.0 mV $s^{-1}$, as described previously [9,19,28]. The sweep was started from the open-circuit potential to the decomposition of the electrolyte. Cyclic voltammetry was performed on the PMT/CNT composite working electrode, with counter and reference electrodes of lithium at a scan rate of 0.5 mV $s^{-1}$. Charge and discharge cycling tests were conducted over a voltage range of 2.5–4.3 V with Toyo battery-test equipment (TOSCAT-3000U).

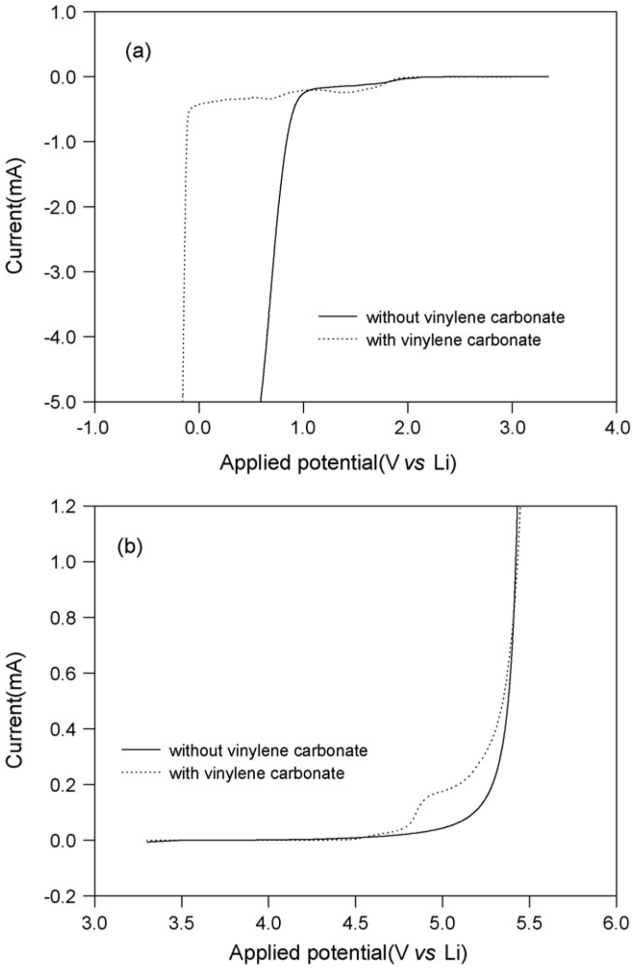

Fig. 1. Linear sweep voltammograms of IL electrolyte (1.0 M $LiBF_4$ in $EMIBF_4$) with and without vinylene carbonate: (a) cathodic scan and (b) anodic scan. Sweep starts from open-circuit potential to decomposition at a scan rate of 1.0 mV $s^{-1}$.

## 3. Results and discussion

Linear sweep voltammetry curves of the IL-based electrolyte without and with vinylene carbonate are shown in Fig. 1(a) and (b), which are obtained for the cathodic and anodic scan, respectively. The cathodic current of the IL electrolyte (1.0 M $LiBF_4$ in $EMIBF_4$) is observed to increase abruptly at around 1.0 V vs. $Li/Li^+$. Because lithium deposition occurs below 0 V, the reduction current may correspond to decomposition of the imidazolium cation. This reduction potential of $EMIBF_4$ is a little lower than the value (1.1 V) reported previously [6]. An oxidative current is observed at around 5.3 V, which corresponds to anodic decomposition of IL electrolyte. In order to prevent the reductive decomposition of $EMIBF_4$, a small amount of vinylene carbonate (VC) was added to the IL electrolyte. As can be seen in Fig. 1(a), the cathodic decomposition of $EMIBF_4$ is hardly observed, but lithium deposition clearly occurs at −0.1 V. The small and broad reduction peak in the potential range of 1.2–1.8 V may be due to reductive decomposition of vinylene carbonate [8,11,29,30], which results in the formation of a surface film that prevents further reductive decomposition of the imidazolium cation of the ionic liquid on the surface of the electrode. Ota et al. [30] have reported that a VC-derived surface film consisted of polymeric species such as poly(vinylene carbonate), the oilgomer of VC and the ring-opening polymer of VC. On sweeping from the open-circuit potential towards positive values, a small oxidative current is observed above 4.7 V for an IL electrolyte containing vinylene carbonate, before decomposition of the IL electrolyte, as shown in Fig. 1(b). The anodic current, which is absent in the undoped IL electrolyte, might originate by the anodic decomposition of vinylene carbonate. From these results, it is understood that adding small amount of vinylene carbonate to the IL electrolyte could increase the cathodic stability of the IL electrolyte, which allows the usage of lithium metal as an anode material for the IL electrolyte.

Morphological analysis of the PMT/CNT composite was carried out through scanning electron micrography, an image is presented in Fig. 2(a). For comparison, the morphology of pure CNT is shown in Fig. 2(b). The diameter of the tubes of CNT is about 20 nm, whereas the fibrils of the PMT/CNT composite have an average diameter of 70 nm. The image of the PMT/CNT composite clearly shows that all the CNTs are quite uniformly coated with poly(3-methylthiophene) and are well dispersed in

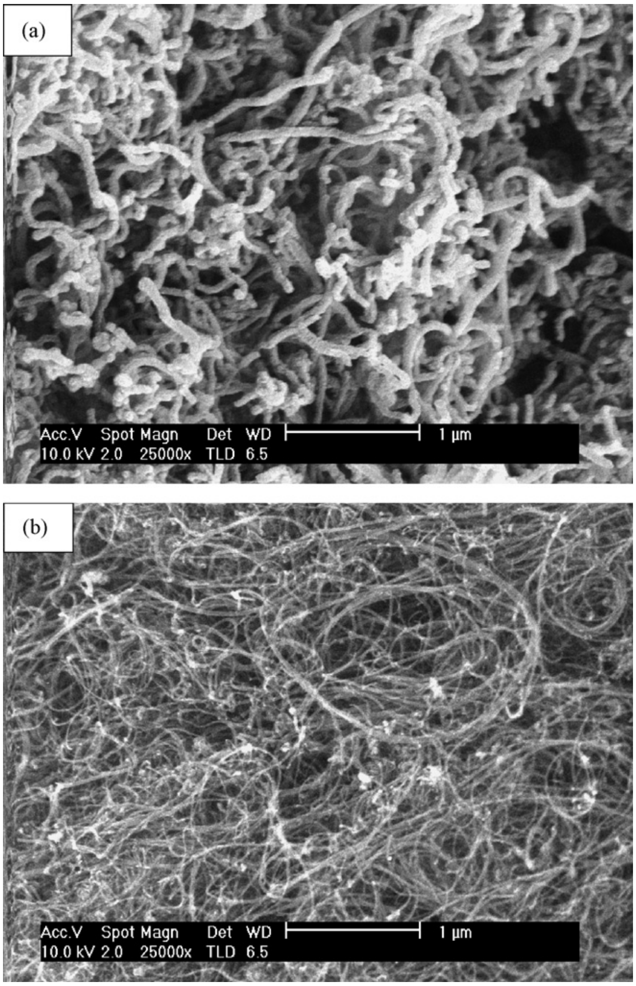

Fig. 2. SEM images of powder samples of (a) PMT/CNT composite and (b) pure CNT.

the composite. This suggests that the CNTs serve as nano-sized framework for the polymerization of 3-methylthiophene. The nanofibrous and porous morphology of the resultant composite is considered to be desirable for a cathode material in lithium batteries because it provides PMT of high-specific-surface area with effective and rapid access of the electrolyte. As previously reported [31,32], CNTs can also offer good mechanical support to the conducting polymer and thereby help to improve its cycleability in rechargeable lithium batteries. Moreover, the CNT ensures good electronic conduction in the composite electrode when the polymer is in an insulating state. From the weight of the composite material, the ratio of PMT to CNT is calculated to be about 70:30 by weight.

In order to identify the oxidation and reduction potentials of the PMT/CNT composite electrode in the IL electrolyte with vinylene carbonate, it was subjected to cyclic voltammetric analysis at a slow scan rate (0.5 mV s$^{-1}$), as shown in Fig. 3. In this figure, the current is normalized by the weight of active material (PMT) in the electrode. The positive currents are for oxidation and the negative currents are for reduction processes. It can be seen that the oxidation reaction occurs at around 3.90 V vs. Li/Li$^+$, while the reduction reaction is in the range of 3.20 and 3.80 V. Such observations of redox processes are similar to those for an unsubstituted polythiophene electrode in a liquid electrolyte [33], and the redox processes are assigned to the doping and de-doping of anions in the polymer during oxidation and reduction, respectively. As expected, these redox potentials are significantly higher than those (3.25 V for oxidation and 2.85 V for reduction) for an analogous PANI/CNT composite electrode in the same ionic liquid electrolyte [22], which suggests that the PMT/CNT electrode can be applied as a high-voltage cathode in lithium metal batteries.

Before subjecting the lithium cells based on the PMT/CNT composite cathode and IL electrolyte to charge–discharge tests, the cells were cycled once at a slow current rate of 0.1 C. This was necessary to allow the formation of the solid-electrolyte interphase (SEI) by the film-forming additive, VC, at the interface of the lithium anode. As mentioned earlier, this SEI plays a role of protecting the imidazolium cations of the IL from reductive decomposition on the surface of lithium anode. After one cycle at the low-current rate, the cells were cycled over a voltage range of 2.5–4.3 V at a constant current equivalent to the 0.2 C rate. Charge–discharge curves of randomly selected cycles for the cells are shown in Fig. 4. These curves show that reversible charge and discharge occurs favourably. The voltage drop in passing from charge to discharge is relatively small, which demonstrates a low-overall resistance of the cell. The cell initially delivered a capacity of 96 mAh g$^{-1}$ based on the weight of active cathode material (PMT). This is similar to the observations of Negatomo and Omoto [34] who reported the specific discharge capacity of PMT in liquid electrolyte ($LiBF_4$ in propylene carbonate and ethylene carbonate) as 97.3 mAh g$^{-1}$ [34]. The discharge capacity of the PMT/CNT cathode is lower than that (139 mAh g$^{-1}$) of a PANI/CNT composite cathode in the same IL electrolyte [22], due to the lower theoretical capacity of PMT (106 mAh g$^{-1}$ at 38% doping) compared with PANI. Despite the lower discharge capacity, the operating voltage of the PMT-based cell is similar to those using transition metal oxides such as $LiCoO_2$, $LiNiO_2$ and $LiMn_2O_4$.

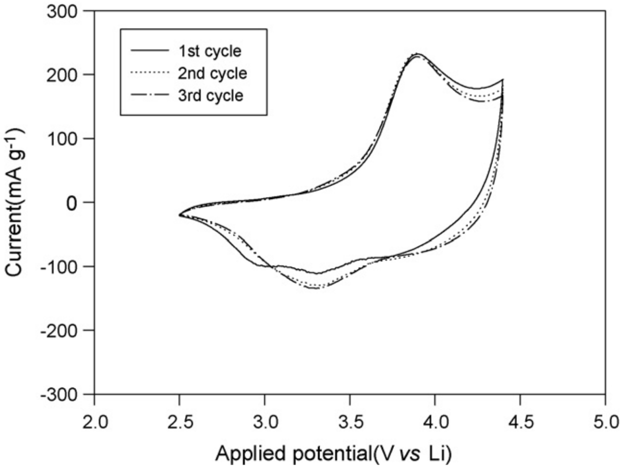

Fig. 3. Cyclic voltammograms of PMT/CNT composite electrode in IL electrolyte containing vinylene carbonate. Scan rate: 0.5 mV s$^{-1}$.

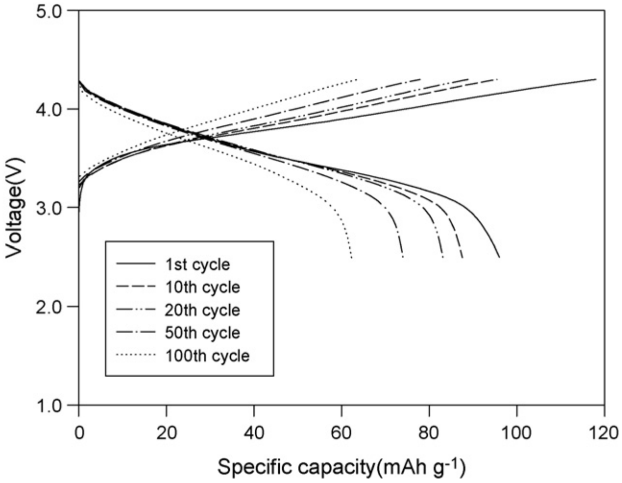

Fig. 4. Charge and discharge curves of lithium metal polymer cells assembled with PMT/CNT composite cathode and IL electrolyte containing vinylene carbonate. Current rates 0.2 C.

Fig. 5 shows plots of charge and discharge capacity as a function of cycle number, which are obtained for IL-based lithium metal polymer cell with and without vinylene carbonate, respectively. The marked differences in the cycle performance of the same cathode material in the two different electrolytes should be noted. The VC-added cell has an initial discharge capacity of 96 mAh g$^{-1}$. This slowly decreases during 100 cycles to a value of 62 mAh g$^{-1}$. The decline in capacity may be related to both deterioration of interfacial contacts at the cathode and oxidative degradation of PMT during cycling. These possibilities will be investigated in the future by optimizing the cathode components and tuning the cut-off potentials. The coulombic efficiency of the cell steadily increases with cycling and reaches over 97.6% at the 100th cycle. The cell without vinylene carbonate gives an initial discharge capacity of 30 mAh g$^{-1}$. Discharge capacity declined rather rapidly until 23 cycles; thereafter it steady decreased up to

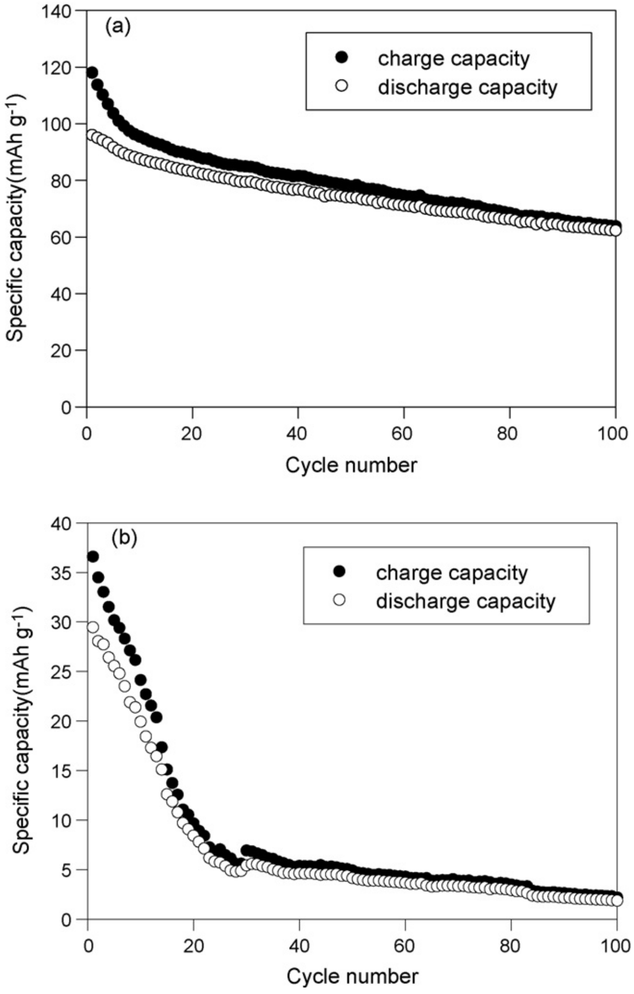

Fig. 5. Charge and discharge capacity of lithium metal polymer cell as function of cycle number obtained for cells assembled with (a) IL electrolyte containing vinylene carbonate and (b) vinylene carbonate-free IL electrolyte. Cycling conducted at 0.2 C rate between 2.5 and 4.3 V.

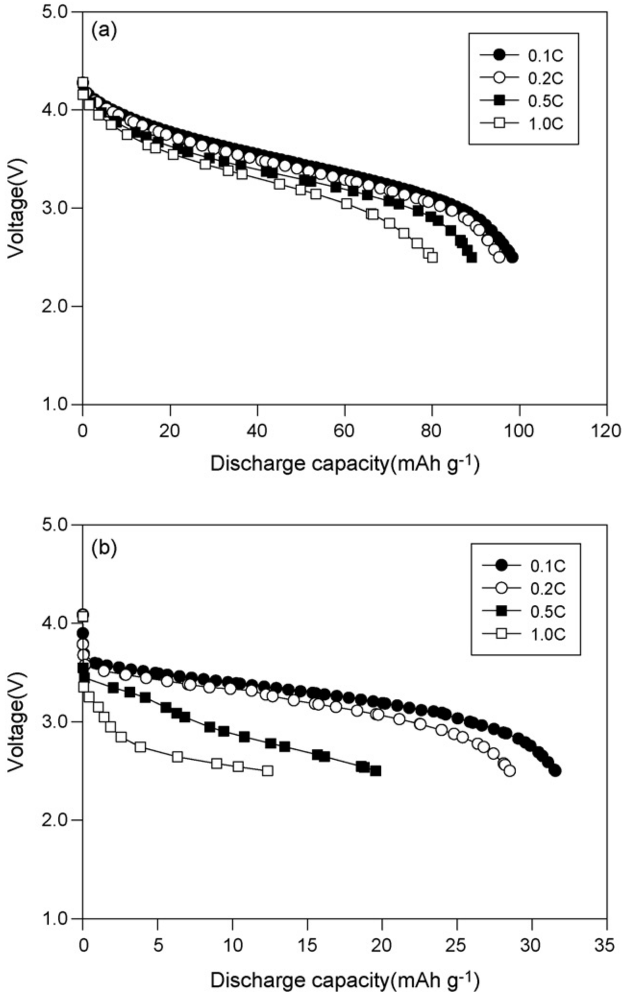

Fig. 6. Discharge profiles of lithium metal polymer cell at different current rates obtained for cells assembled with (a) IL electrolyte containing vinylene carbonate and (b) vinylene carbonate-free IL electrolyte.

100 cycles to a value of 2 mAh $g^{-1}$. The coulombic efficiency is 85.5% at the 100th cycle. As mentioned earlier, the VC additive used in the IL-based cell forms a thin and homogeneous SEI film, which offers efficient protection to the imidazolium cation of the IL so that it is not decomposed on the lithium anode surface. Thus, the cell with the VC additive results in higher capacity and better cycleability. It has been reported that most of the VC added into the electrolyte is used up in SEI formation and hence the incombustibility of IL-based cells is not at all spoiled by the addition of a small quantity of organic additive in the cell [5].

The rate capabilities of the lithium metal polymer cells assembled with a PMT/CNT composite cathode and an IL electrolyte were evaluated. In order to induce formation of the SEI on the surface of the lithium electrode, the cells were initially subjected to two cycles at the 0.1 C rate prior to rate capability tests. The discharge curves of the cells at different current rates are given in Fig. 6. With both types of cell, the average discharge voltage and capacity decrease on increasing the current rate from 0.1 to 1.0 C for both cells. For the cell containing VC, the discharge capacity is about 80 mAh $g^{-1}$ at the 1.0 C rate, which corresponds to 83% with respect to the capacity at the 0.1 C rate. By contrast, the discharge capacity of the cell assembled with VC-free IL electrolyte is very small (12 mAh $g^{-1}$) at the 1.0 C rate. This marked decline in performance may be due to the formation of a poorly conductive SEI on the anode surface, which results from the reductive decomposition of the imidazolium cation of the ionic liquid. Further studies are being conducted in order to improve the cycleability and high-rate performances of lithium metal polymer cells assembled with a PMT/CNT composite cathode and an IL electrolyte by using different ILs, optimizing the quantity of SEI film-forming agent, and tuning the cut-off potentials.

## 4. Conclusions

A PMT/CNT composite has been synthesized and employed as a cathode material in a lithium metal polymer cell assembled with an IL electrolyte. In this cell, VC is added in order to form a SEI layer and protect the imidazolium cations of the IL from reductive decomposition on the surface of the lithium anode. The

VC-added cell yields higher discharge capacity and better capacity retention than the cell without VC. The cell gives an initial discharge capacity of 96 mAh $g^{-1}$ with relatively good capacity retention and shows a discharge capacity of 80 mAh $g^{-1}$ at the 1.0 C rate. Although the specific capacity of the cell is relatively small, the lithium metal polymer cells assembled with a non-flammable IL electrolyte and a PMT/CNT composite electrode may be potential candidates for high-voltage lithium polymer batteries with enhanced safety.

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