

An improved electrolyte for the LiFePO_4 cathode working in a wide temperature range

S.S. Zhang*, K. Xu, T.R. Jow

U.S. Army Research Laboratory, Adelphi, MD 20783-1197, USA

Received 3 October 2005; received in revised form 30 October 2005; accepted 2 November 2005

Available online 27 December 2005

Abstract

A LiBF_4 – LiBOB (lithium bis(oxalato)borate) salt mixture was used to formulate an electrolyte for the operation of a LiFePO_4 cathode over a wide temperature range (-50 to 80°C) by employing a solvent mixture of 1:1:3 (wt.) propylene carbonate (PC)/ethylene carbonate (EC)/ethylmethyl carbonate (EMC). In comparison with the ionic conductivity of a single salt electrolyte, LiBF_4 electrolyte has a higher conductivity below -10°C while the LiBOB electrolyte is higher above -10°C . For cell performance, LiBF_4 cell has a better low temperature performance and a higher power capability, but it cannot survive above 60°C . In contrast, the LiBOB cell performs very well at high temperature even up to 90°C , but it fails to perform below -40°C . We found that the temperature performance of $\text{Li}/\text{LiFePO}_4$ cells could be optimized by using a LiBF_4 – LiBOB salt mixture. At 1C and at -50°C , for example, a $\text{Li}/\text{LiFePO}_4$ cell using 90:10 (in mole) LiBF_4 – LiBOB salt mixture could provide up to $\sim 30\%$ of capacity at $\sim 3.0\text{V}$ and it still could be cycled at 90°C . In addition, we observed and explained an opposite correlation between the ionic conductivity of the electrolyte and the power capability of the cell. That is, the LiBF_4 cell at 20°C discharges at a higher plateau voltage than the LiBOB cell, whereas the LiBF_4 electrolyte has a lower ionic conductivity.

Published by Elsevier B.V.

Keywords: LiFePO_4 ; LiBF_4 ; Lithium bis(oxalato)borate; Ionic conductivity; Low temperature performance; High temperature performance

1. Introduction

In 1997, Padhi et al. [1] first proposed olivine LiFePO_4 as a new cathode material for rechargeable lithium batteries. The most significant advantages of this material are its excellent stability in the charged state and it is environmentally benign, which makes it attractive for the development of heavy-duty lithium-ion batteries such as those for use in electric vehicles and hybrid electric vehicles. The main limitation of this material is its poor rate capability, which is attributed to the inherently low electronic conductivity and slow kinetics of lithium ion diffusion through the LiFePO_4 – FePO_4 interfaces [2,3]. Two approaches have been proposed to solve this problem: one is to enhance bulk electronic conductivity of the LiFePO_4 particles by cation doping [4–6] and the other is to increase surface electronic conductivity of the particles by making a LiFePO_4 –C composite or so-called “carbon-coating” [7–9]. However,

few papers are concerned with the improvement on this cathode material by modifying formulation of the electrolytes [10,11].

More recently, Amine et al. reported [10] that upon long-term cycling, Fe(II) ions in the LiFePO_4 dissolve into the LiPF_6 -based electrolyte and the dissolved Fe(II) ions migrate toward the anode and reductively deposit there. This not only reduces capacity but also the performance of the anode. Fortunately, such dissolutions can be effectively suppressed in the LiBOB -based electrolyte because Fe(II) ions and LiBOB react with each other to form insoluble products that cover the surface of LiFePO_4 particles and hence prevent further dissolution. Other advantages of the LiBOB -based electrolyte are known to include (1) enhanced safety of the LiFePO_4 cathode [11] and graphite anode [12], respectively; (2) participating in the formation of a solid electrolyte interface (SEI) and stabilizing the SEI on the graphite surface [13]; (3) allowing the lithium-ion cell to operate at high temperature [14]. Barriers for the application of LiBOB in Li-ion cells have been recognized as [15] (1) low solubility in linear alkyl carbonates and high viscosity of its solution, which both limit low temperature and high current rate

* Corresponding author. Tel.: +1 301 394 0981; fax: +1 301 394 0273.
E-mail address: szhang@arl.army.mil (S.S. Zhang).

performance of the Li-ion cells; (2) oxidative instability at high potentials (>4.2 V), which could result in gas generation; and (3) extreme sensitivity to moisture, which requires an extremely dry solvent.

On the other hand, we previously found [16–19] that a Li-ion cell with a LiBF_4 -based electrolyte, in addition to having better high temperature performance, has excellent low temperature performance. This is because at low temperature the LiBF_4 electrolyte has low viscosity and more importantly such a cell presents a much lower charge-transfer resistance, which is believed to favor cell reactions on the electrolyte–electrolyte interface [17,18]. In spite of many other advantages of LiBF_4 over LiPF_6 , such as better thermal stability and less sensitivity to water, application of LiBF_4 in Li-ion cells is limited mainly by its poor ability in facilitating formation of a stable SEI on the graphite surface. Based on the above knowledge and on the fact that LiFePO_4 cathode operates at a flat voltage of ~ 3.4 V versus Li^+/Li , which allows LiBOB to be stable, we speculated that an optimized electrolyte can be formulated for the LiFePO_4 cathode by using a LiBF_4 –LiBOB salt mixture. In this work, we aimed at employing a 1:1:3 PC/EC/EMC mixed solvent to formulate the electrolyte that enables LiFePO_4 cathode to operate in a wide temperature range (-40 to 80 °C), while still having good energy and power density. In the present work we evaluate and discuss the cycling performance of the Li/LiFePO₄ cell using the formulated electrolyte in terms of low temperature, high current, and high temperature.

2. Experimental

LiBF_4 (Stella Chemifa Corp.) and LiBOB (Chemmetal, Germany) were dried at 100 °C under vacuum for 8 h before use. Propylene carbonate (PC), ethylene carbonate (EC), and ethylmethyl carbonate (EMC, all from Ferro Chemical) were dried in sequence using 3 Å molecular sieves and neutral alumina. In an argon-filled glove box, four electrolytes with a general composition of 1.0 m $(1-x)\text{LiBF}_4-x\text{LiBOB}$ 1:1:3 (wt.) PC/EC/EMC, where $x=0, 0.1, 0.5,$ and 1.0 , respectively, were prepared. For the one with $x=1$, however, only 0.8 m solution was prepared due to the limited solubility of LiBOB in the solvent mixture.

LiFePO_4 -C (95:5 wt.) composite was synthesized by the solid-state reaction of $\text{FeC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ (99%, Aldrich), LiH_2PO_4 ($>99.99\%$, Aldrich), and black pearls 2000 (a highly conductive carbon). The detailed synthesis procedures were described elsewhere [9]. Electrode film with a load of 10 ± 1 mg cm^{-2} and a composite of 80% cathode active material, 15% carbon black, and 5% binder was coated onto an aluminum foil by using poly(acrylonitrile-co-methyl methacrylate) as the binder and *N*-methylpyrrolidone as the solvent. After drying in air, the resulting electrode sheet was cut into small discs with an area of 1.27 cm^2 and dried at 120 °C for 8 h under vacuum before use. In the glove-box, BR2335-size Li/LiFePO₄ button cells were assembled using Celgard® 2500 membrane as the separator and filled with 80 μL of liquid electrolyte.

A Tenney Environmental Oven Series 942 was used to provide a constant temperature environment for the test. Solartron SI 1287 Electrochemical Interface and SI 1260 Impedance/Gain-Phase Analyzer were employed to measure impedance and to perform electrochemical measurement. Ionic conductivity of the liquid electrolytes was determined by measuring the impedance of a two-electrode cell inserted in the liquid electrolyte. Passivation behavior of the cathode substrate (aluminum) in electrolytic solution was studied by a linear sweep voltammetry (LSV) experiment, in which a newly scratched aluminum wire was adapted as the working electrode and lithium foils as the counter and reference electrodes, respectively. An aluminum wire (diameter = 1.0 mm, purity = 99.999% , Aldrich) was wrapped with a thermal shrinkable Teflon® tube by leaving a 1.0 cm length exposed to air and then heating the tube to shrink. Each aluminum wire was scanned twice from the open circuit potential (OCP) to 6.5 V versus Li^+/Li at a sweeping rate of 5 mV s^{-1} and the I - E response was recorded.

Cycling test of the Li/LiFePO₄ button cells was performed using a Maccor Series 4000 tester between 2.0 and 4.2 V. Current density for charge and discharge was expressed as the C rate, which was calculated by assuming specific capacity of the LiFePO_4 -C composite to be 170 mA h g^{-1} . More detailed testing conditions are described either in text or in figure caption.

3. Results and discussion

3.1. Selection of solvent system

In this work, a PC/EC/EMC ternary solvent mixture was chosen to formulate the Li-ion electrolyte with a wide operating temperature range. This selection was based on the combined merits of freezing temperature, wettability with Celgard® membrane, and the electrochemical characteristics related to SEI formation on the graphite surface. Table 1 compares the main features of the common carbonate solvents used in Li-ion cells. It has been known that EC is an essential component for the formation of a stable SEI. However, the content of EC in low temperature electrolyte is limited due to its high melting point (36.4 °C), poor miscibility with linear alkyl carbonates, and inability to wet Celgard® membrane. DMC has a high melting point (4.6 °C), whereas DEC is chemically unstable with lithiated graphite and has least miscibility with EC. Therefore, DMC and DEC would not be the perfect solvent for the low temperature electrolyte. Considering the overall characteristics of the melting point, viscosity, wettability to the separator, and SEI formation, we selected EMC as the major component to develop the low temperature electrolyte. The roles of EMC in the electrolyte include diluting viscosity, enhancing wettability, and participating SEI formation. In our previous work [20], we found that EC/EMC binary solvent is not enough to form low freezing point because of their poor miscibility. Therefore, we used a small amount of PC as the third component to lower freezing point of the electrolyte although PC is unable to participate into SEI formation [21].

Table 1
Comparison of the common carbonate solvents used in Li-ion cells

	mp (°C)	Advantage	Drawback
EC	36.4	High polarity and an essential component for SEI formation of graphite	High mp and poor miscibility with linear alkyl carbonates, both of which results in a high freezing point
PC	−48.8	Low mp and excellent miscibility with EC, which lowers freezing temperature	High viscosity and poor SEI formation due to co-intercalating and decomposing
DMC	4.6	Excellent wettability with Celgard membrane® and good for SEI formation	High mp and poor miscibility with EC
DEC	−74.3	Excellent wettability with Celgard membrane® and lowest mp	Poor chemical stability with lithiated graphite and least miscibility with EC
EMC	−53	Best combination of the mp, wettability, and cell performance	Insufficient miscibility with EC to form very low freezing point

Note: All mp data was cited from ref. [20].

Considering the combined factors of freezing point and SEI formation, we here selected a 1:1:3 PC/EC/EMC ternary solvent system to formulate the low temperature electrolyte for the LiFePO₄-based Li-ion cells. This solvent system has been known not only to have a low freezing point but also to have very good super-cooling ability. To examine the freezing of such electrolytes, we stored a 1.0 m LiBF₄ 1:1:3 PC/EC/EMC solution in a −40 °C oven for two days and found that the solution remained in a single liquid phase without salt precipitating and EC crystallizing out of the solution.

3.2. Ionic conductivity and Al passivation behavior

Fig. 1 compares $\log \sigma - 1/T$ plots of the electrolytes having a general composition of 1.0 m (1-x)LiBF₄-xLiBOB 1:1:3 PC/EC/EMC, where x=0, 0.1, 0.5, and 1 (0.8 m for the case of x=1 due to the limited solubility of LiBOB in the solvent). In general, all these plots are smooth, which suggests no phase change occurring even down to the lowest testing temperature (−50 °C). It is shown that the $\log \sigma - 1/T$ plots for these having x=0 and 1.0, respectively, are crossed at −10 °C. Above

−10 °C, LiBOB electrolyte (x=1) has higher ionic conductivity than LiBF₄ electrolyte (x=0), whereas the case reverses as the temperature is lower than −10 °C. This phenomenon is attributed to the combined effect of salt dissociation and solution viscosity. Compared with LiBF₄, LiBOB is more easily dissociated in the solution because of its larger anion, which favors weakening ion-pairings. When ion dissociation is the predominant contribution to the ionic conductivity, LiBOB electrolyte exhibits higher ionic conductivity. At low temperature, the increased viscosity of the solution could become the main barrier to the ionic conduction. In such a case, ionic conductivity of LiBOB electrolyte is lower as viscosity of the LiBOB solution is increased much faster than that of LiBF₄ solution with decreasing of the temperature [22]. Ionic conductivity of the electrolytes with x=0.1 and 0.5 are found to lie between those of the LiBF₄ and LiBOB electrolytes, demonstrating an average effect (Fig. 1).

Fig. 2 displays I-E response of the initial two cycles for a newly scratched aluminum wire inserted in the electrolyte with x=0 and 1, respectively. In the first cycle, the I-E response in both electrolytes exhibits an S-shape profile and furthermore

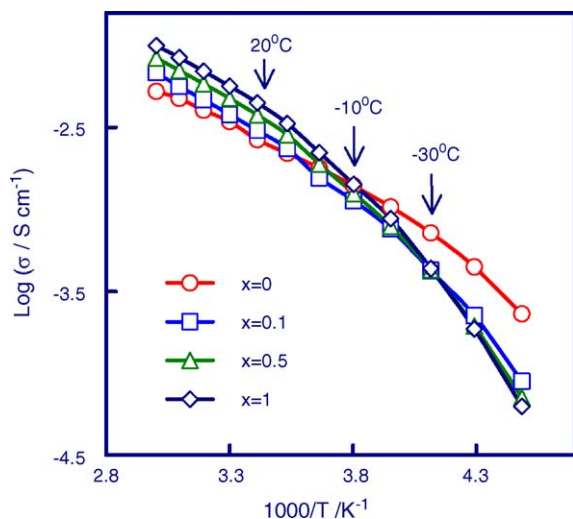


Fig. 1. Ionic conductivities of the electrolytic solutions with a composition of 1.0 m (1-x)LiBF₄-xLiBOB 1:1:3 PC/EC/EMC (0.8 m for x=1).

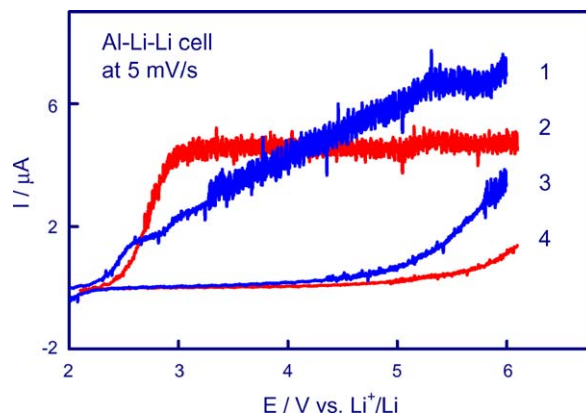


Fig. 2. I-E response of a newly scratched aluminum wire in an electrolytic solution of (a) 0.8 m LiBOB 1:1:3 PC/EC/EMC and (b) 1.0 m LiBF₄ 1:1:3 PC/EC/EMC, respectively, which was recorded at a sweeping rate of 5 mV s⁻¹. (1) First sweep in (a), (2) first sweep in (b), (3) second sweep in (a), and (4) second sweep in (b).

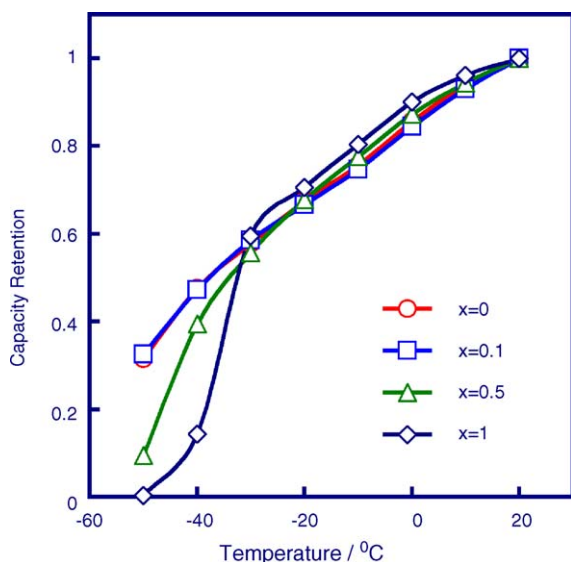


Fig. 3. Effect of salt ratio on the low temperature performance of Li/LiFePO₄ cells with an electrolyte of 1.0 m (1 - x)LiBF₄-xLiBOB 1:1:3 PC/EC/EMC (0.8 m for x = 1), in which the cells were cycled at 1C and the capacity retention is defined as the ratio of discharge capacity at a specific temperature to the capacity at 20 °C.

the currents throughout the scanning potential range are very low (only a few μA , see curves 1 and 2). These are characteristics of the highly effective passivation of aluminum. The currents in the second cycle are significantly lower than those observed in the first cycle (see curves 3 and 4), which is another evidence for the effective passivation accrued in the previous cycle. The observations above verify that both LiBF₄ and LiBOB are electrochemically compatible with the current collector material (aluminum) of the cathode. In addition, it should be noted that in the potential range of higher than 4.5 V, the currents with LiBOB electrolyte (curve 3) are slightly higher than those with LiBF₄ electrolyte (curve 4). This phenomenon may be associated with the slow oxidization of LiBOB at high potentials.

3.3. Cell performance at low temperature

Fig. 3 compares the capacity retention of the Li/LiFePO₄ cells with different electrolytes when cycled at 1C and at -30 °C, in which the capacity retention is expressed as the ratio of discharge capacity at a specific temperature to that at 20 °C. It is shown that above -30 °C, all these cells have the similar retentions. In other words, there is no direct correlation between capacity retention and electrolytic ionic conductivity. When the temperature goes down below -30 °C, the capacity difference becomes very obvious. For example, at -50 °C LiBF₄ cell (x = 0) still remains over 30% capacity while LiBOB cell (x = 1) entirely loses its cycling ability. However, the cell with x = 0.1 retains nearly the same capacity as the LiBF₄ cell over all the testing temperature range. This reveals that addition of a small amount of LiBOB into the LiBF₄ electrolyte has no adverse impact on the energy capacity (specific capacity) of Li/LiFePO₄ cell at low temperature.

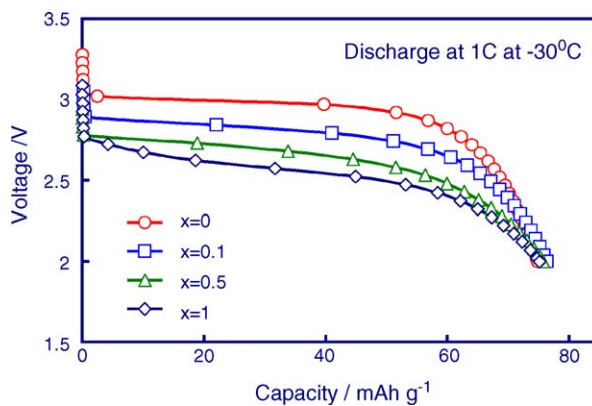


Fig. 4. Effect of salt ratio on the discharge voltage of Li/LiFePO₄ cells, which was measured at 1C by charging the cell at 20 °C and then discharging at -30 °C.

Although the cells with different LiBF₄-LiBOB ratio demonstrated similar energy capacities at -30 °C (Fig. 3), their discharge voltage (power capability) was significantly different (Fig. 4). As Fig. 4 shows, the discharge voltage of the cells decreases with the addition of LiBOB into LiBF₄. To find the correlation between cell power capability and electrolytic ionic conductivity, we compared ionic conductivity of the electrolytes shown in Fig. 1. Except for LiBF₄ electrolyte (x = 1), all other three electrolytes (x = 0.1, 0.5, and 1.0, respectively) have nearly the same conductivity at -30 °C. This fact suggests that the electrolytic ionic conductivity could not be the only factor to determine cell power capability. We previously found [17,18] that at low temperature, LiBF₄ Li-ion cells show higher energy and power capability than the counterpart LiPF₆ cell in spite of the relatively low ionic conductivity of the LiBF₄ electrolyte. This merit is attributed to the lower charge-transfer resistance of the LiBF₄ Li-ion cell. We believe that the same reason observed in Li-ion cells is applicable to the present Li/LiFePO₄ cells.

Fig. 5 compares discharging curves of the Li/LiFePO₄ cell with an electrolyte of x = 0.1 at different temperatures. A general trend is that with decreasing of the temperature, both energy and

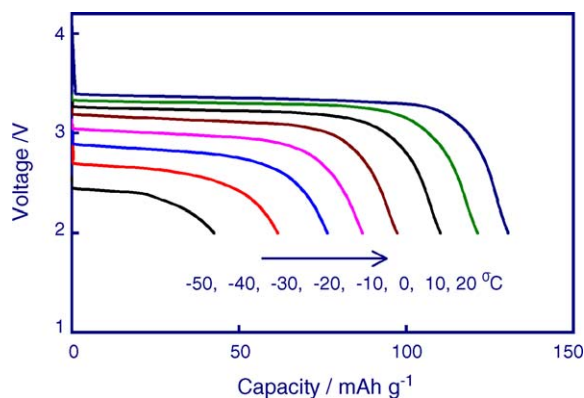


Fig. 5. Discharge curves of Li/LiFePO₄ cell with a 1.0 m (0.9LiBF₄-0.1LiBOB) 1:1:3 PC/EC/EMC electrolyte, which were recorded at 1C by charging the cell at 20 °C and then discharging at a specific temperature.

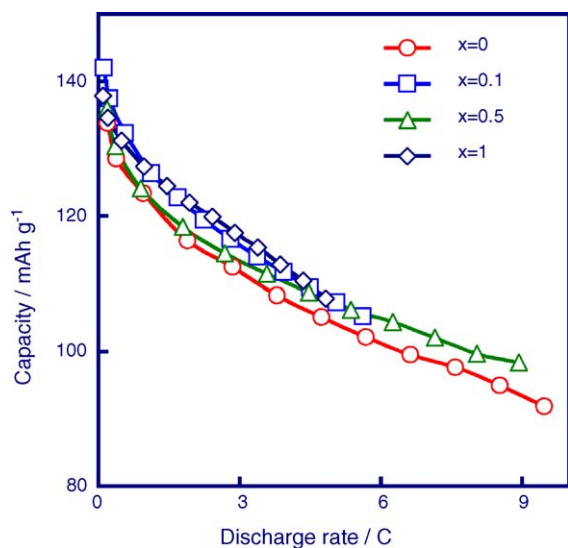


Fig. 6. Change of discharge capacities with the current rate for the Li/LiFePO₄ cells using 1.0 m (1-x)LiBF₄-xLiBOB 1:1:3 PC/EC/EMC electrolyte (0.8 m for x=1), in which the tests were conducted at 20 °C by charging the cell at 1C and then discharging at a specific current rate.

power capability are reduced. This phenomenon is attributed to the decreased ionic conductivity of the electrolyte and to the slowed-down kinetics of cell reactions, which are suffered at low temperatures. In addition, Fig. 5 shows that the cell deliveries up to 100 mA h g⁻¹ capacity with a plateau voltage of higher than 3.0 V when being discharged at 1C and at -10 °C.

3.4. Cell performance at high current rate

The effect of the salt ratio on the discharge capacity at various current rates for the Li/LiFePO₄ cells is shown in Fig. 6. At a low current rate (0.05C), all these cells deliver about 140 mA h g⁻¹ of full capacity, i.e., about 82% of theoretical specific capacity of the pure LiFePO₄ cathode. The discharge capacity is gradually decreased with an increase in the current rate. However, there is no distinct correlation between the discharge capacity and salt ratio except for the LiBF₄ cell that has a slightly poorer rate capability as compared with the other cells. This may be associated with the lowest ionic conductivity of the LiBF₄ electrolyte at the testing temperature (see Fig. 1).

Fig. 7 presents the impact of salt ratio on discharge voltage (power capability) of the Li/LiFePO₄ cells. Although these cells discharge nearly the same capacity (about 105 mA h g⁻¹) at 5C, the discharge voltage is decreased with increasing of LiBOB ratio in the salt mixture. For example, at 20 °C LiBF₄ electrolyte has lowest ionic conductivity (Fig. 1), while the cell with it presents the highest plateau voltage (Fig. 7). These phenomena are consistent with the results shown in Fig. 4, however, they cannot be interpreted in terms of the ionic conductivity of the electrolytes. The total impedance of a cell is generally composed of the bulk resistance, surface layer impedance, charge-transfer impedance and diffusion-related impedance. For the LiBF₄ cell, we found it has a lower charge-transfer resistance, especially at low temperature [17,18]. We believe that the good performance

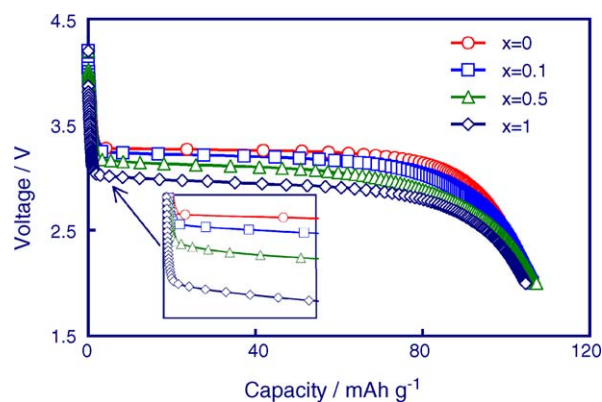


Fig. 7. Effect of salt ratio on the discharge curves of the Li/LiFePO₄ cells cycled at 5C and at 20 °C.

of the LiBF₄ cell at low temperature (Fig. 4) and at high current rate (Fig. 7) must be associated with the low charge-transfer resistance of the LiBF₄ cell.

Discharging curves at different current rates for the Li/LiFePO₄ cell with an electrolyte of x=0.1 are plotted in Fig. 8, which indicates very good power capability of the LiBF₄-LiBOB salt mixture cell. At 0.1C, the cell delivers 142 mA h g⁻¹ capacity with an plateau voltage of 3.4 V. When the current is increased to 5C, the cell still remains up to 107 mA h g⁻¹ capacity and 3.1 V plateau voltage. From the results above, we concluded that the LiBF₄ electrolyte is very good for promoting low temperature and high current rate performance of the Li/LiFePO₄ cell, and that most of the advantages of the LiBF₄ cell can be retained in the LiBF₄-LiBOB salt mixture cell when the molar ratio of LiBOB is less than 0.1.

3.5. Cell performance at high temperature

Cycling performance of the Li/LiFePO₄ cells at high temperature was evaluated. In this experiment, the cells were run for 50 cycles at each temperature and then the temperature was raised by 10 °C for the next cycling test. The discharge capacity results

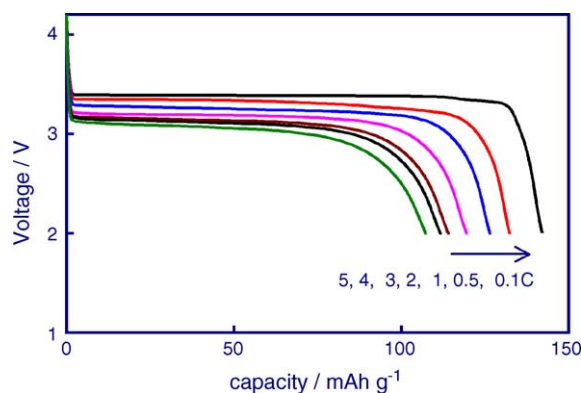


Fig. 8. Discharge curves of the Li/LiFePO₄ cells with a 1.0 m (0.9LiBF₄-0.1LiBOB) 1:1:3 PC/EC/EMC electrolyte, which were recorded at 20 °C by charging the cell at 1C and then discharging at a specific current rate.

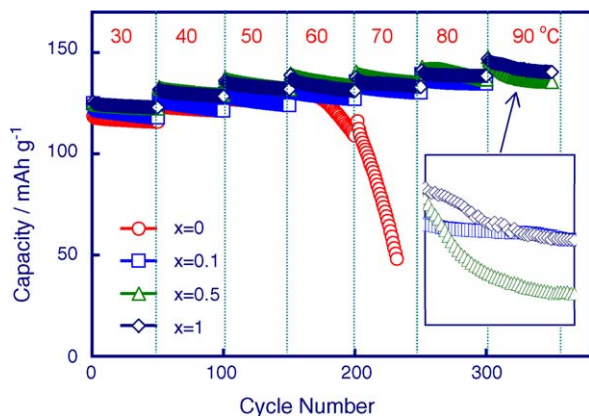


Fig. 9. Comparison of the cycling performance at high temperature for the Li/LiFePO₄ cells with different electrolytes, in which the cells were charged and discharged at 1C.

are summarized in Fig. 9, which shows that LiBOB cell can be cycled at 80 °C without visible fading. Even at 90 °C, the LiBOB cell still could be cycled with a slightly accelerated capacity fading. The excellent high temperature performance of the LiBOB cell is attributed to these two advantages related to the LiBOB electrolyte: (1) excellent thermal stability of LiBOB salt [11,12] and (2) substantially suppressed dissolution of Fe(II) ions from LiFePO₄ into LiBOB electrolyte [10]. By contrast, the LiBF₄ cell is unable to survive above 60 °C most likely because of the accelerated dissolution of Fe(II) ions at the high temperature although the LiBF₄ electrolyte itself is stable at such temperatures [16]. Fortunately, the high temperature performance of the LiBF₄ cell can be significantly improved by addition of a small amount of LiBOB, while the cell still provides good cycling performance at low temperature and at high current rates.

4. Conclusions

In conclusion, the LiBF₄ cell is capable of providing good cycling performance at low temperature and at a high current rate, while it is unable to survive above 60 °C. By contrast, the LiBOB cell provides excellent cyclability at high temperature, while it suffers poor power capability (low plateau voltage) at low temperature and at a high current rate. The cycling performance of the Li/LiFePO₄ cell at low temperature, high

current rate, and high temperature can be improved by using a LiBF₄–LiBOB salt mixture. However, the optimized performance is achieved only when a molar ratio of LiBOB in the salt mixture is less than 0.1. On the other hand, it was found that the power capability of the Li/LiFePO₄ cell is more dependent on the charge-transfer resistance, rather than the electrolytic ionic conductivity. Featuring a low charge-transfer resistance, the LiBF₄ cell is able to provide good power capability although the ionic conductivity of LiBF₄ electrolyte is relatively low above –10 °C.

References

- [1] A.K. Padhi, K.S. Nanjundaswamy, C. Masquelier, S. Okada, J.B. Goodenough, *J. Electrochem. Soc.* 144 (1997) 1609.
- [2] A.S. Andersson, J.O. Thomas, B. Kalska, L. Haggstrom, *Electrochem. Solid-State Lett.* 3 (2000) 66.
- [3] A.S. Andersson, J.O. Thomas, *J. Power Sources* 97–98 (2001) 498.
- [4] S.Y. Chung, J.T. Bloking, Y.M. Chiang, *Nat. Mater.* 1 (2002) 123.
- [5] J. Allen, K. Xu, S.S. Zhang, M. Ding, T.R. Jow, 204th ECS Meeting Abstracts, No. 347, Orlando, FL, 12–17 October 2003.
- [6] S. Shi, L. Liu, C. Ouyang, D.S. Wang, Z. Wang, L. Chen, X. Huang, *Phys. Rev. B* 68 (2003) 195108.
- [7] H. Huang, S.C. Yin, L.F. Nazar, *Electrochem. Solid-State Lett.* 4 (2001) A170.
- [8] Z. Chen, J.R. Dahn, *J. Electrochem. Soc.* 149 (2002) A1184.
- [9] S.S. Zhang, J.L. Allen, K. Xu, T.R. Jow, *J. Power Sources* 147 (2005) 234.
- [10] K. Amine, J. Liu, I. Belharouak, *Electrochem. Commun.* 7 (2005) 669.
- [11] J. Jiang, J.R. Dahn, *Electrochem. Commun.* 6 (2004) 724.
- [12] J. Jiang, J.R. Dahn, *Electrochem. Solid-State Lett.* 6 (2003) A180.
- [13] K. Xu, S.S. Zhang, B.A. Poese, T.R. Jow, *Electrochem. Solid-State Lett.* 5 (2002) A259.
- [14] K. Xu, S.S. Zhang, T.R. Jow, W. Xu, C.A. Angell, *Electrochem. Solid-State Lett.* 5 (2002) A26.
- [15] K. Xu, S.S. Zhang, U. Lee, J.L. Allen, T.R. Jow, *J. Power Sources* 146 (2005) 79.
- [16] S.S. Zhang, K. Xu, T.R. Jow, *J. Electrochem. Soc.* 149 (2002) A586.
- [17] S.S. Zhang, K. Xu, T.R. Jow, *Electrochem. Commun.* 4 (2002) 928.
- [18] S.S. Zhang, K. Xu, T.R. Jow, *J. Solid State Electrochem.* 7 (2003) 147.
- [19] S.S. Zhang, K. Xu, T.R. Jow, 205th ECS Meeting Abstracts, No. 82, San Antonio, TX, 9–14 May 2004.
- [20] M.S. Ding, K. Xu, S.S. Zhang, T.R. Jow, *J. Electrochem. Soc.* 148 (2001) A299.
- [21] S.S. Zhang, K. Xu, J.L. Allen, T.R. Jow, *J. Power Sources* 110 (2002) 216.
- [22] M.S. Ding, K. Xu, T.R. Jow, *J. Electrochem. Soc.* 152 (2005) A132.