JOURNAL OF APPLIED ELECTROCHEMISTRY **27** (1997) 902–907

Influence of the composition of ternary mixed solvent electrolytes on the properties of rechargeable lithium cells

S. -I. TOBISHIMA, K. HAYASHI, Y. NEMOTO, J. -I. YAMAKI

NTT Interdisciplinary Research Laboratories, Nippon Telegraph and Telephone Corporation,
Tokai-mura, Ibaraki-ken, 319-11, Japan

Received 8 July 1996; revised 15 October 1996

A cell system consisting of an amorphous (a-)V_2O_5–P_2O_5 (95:5 in molar ratio) cathode, a lithium metal anode and an organic electrolyte by fabricating an AA-size prototype cell has been studied. The influence of the composition of ternary mixed solvent electrolytes comprising ethylene carbonate (EC), propylene carbonate (PC) and 2-methyltetrahydrofuran (2MeTHF) on the properties of AA-size Li / a-V_2O_5–P_2O_5 cells is examined. The goals are to improve the cycle life of the cell while ensuring its safety and controlling its cost. The electrolyte composition is varied by changing the solvent mixing ratio, and the lithium salt concentration, and by using different kinds of solute. The cell performance examined includes cathode utilization (cell capacity) during low temperature operation, and charge–discharge cycle life. Heating abuse tests were also carried out at 130 °C on the Li / a–V_2O_5–P_2O_5 cells. The best electrolyte composition was found to be 1.15 M $LiAsF_6$–EC / PC / 2MeTHF (15:70:15) from the overall results on cycle life, capacity, safety and cost.

---

1. Introduction

Rechargeable cells with Li metal anodes are attractive because theoretically they have a higher energy density than Li ion cells containing Li inserted carbon anodes which have already been produced commercially by several manufacturers [1]. With the Li metal battery system, the most important problems to be solved are those related to cycle life and safety. These problems are closely connected with the selection of a suitable electrolyte for this cell system. The composition, including the solvent mixing ratio and the salt concentration, should be optimized and the electrolyte materials carefully chosen, when determining a suitable electrolyte.

A cell system consisting of an amorphous (a-)V_2O_5–P_2O_5 cathode, a lithium (Li) metal anode and an organic electrolyte, by fabricating an AA-size cell has been studied [2]. In a previous study [3], we reported the influence of the mixing ratio of ethylene carbonate (EC) and propylene carbonate (PC) solvents, in EC/PC binary mixed solvent systems incorporating $LiAsF_6$ as the solute, on the rate capability, cycle life and safety of Li/a–V_2O_5–P_2O_5 cells. An EC content of 10–30 vol% was found to exhibit the maximum cycle life with relatively higher capacity. This result is believed to be related to the interaction between EC and $Li_xV_2O_5$ [3, 4]. The influence of the addition of a small amount of 2-methyl-tetrahydrofuran (2MeTHF) to an EC/PC binary mixed system on AA Li/a-V_2O_5–P_2O_5 cell properties has also been briefly examined [5]. Based on such considerations as safety, cost and ease of handling in a manufacturing plant, a lower 2MeTHF content would be better. This is because 2MeTHF has a low flash point of −11 °C, a low boiling point of 79 °C, it is easily oxidized leading to the formation of dangerous peroxide, it is hydroscopic and its cost is about five times that of EC or PC [6]. In addition, an investigation of the thermal stability of organic electrolytes by accelerating rate calorimetry (ARC) [7] indicated that, in terms of safety, the preferable 2MeTHF content of an organic electrolyte system is less than 15 vol%.

In this paper, we describe a study of the influence of the composition of EC/PC/2MeTHF ternary mixed solvent electrolyte on the properties of AA-size Li/a-V_2O_5–P_2O_5 cells with a view to improving the cycle life of the cell while ensuring its safety and controlling its cost. In this work, the 2MeTHF content is fixed at 15 vol% based on the results of previous studies related to safety and cost [5, 7]. The material cost of EC/PC/2MeTHF (15 vol% 2MeTHF) is about twice that of the EC/PC binary mixed system. Electrolyte composition was varied by changing the mixing ratio of the EC and PC solvents (EC:PC is 5:80–42.5:42.5) and the $LiAsF_6$ concentration (0.75–1.5 mol dm^{-3}), and by using different solute ($LiPF_6$ or $LiN(CF_3SO_2)_2$). The cell properties include capacity and cycle life. A heating abuse test was also carried out.

2. Experimental details

2.1. *Electrolytes*

Electrolyte solutions were prepared as described in previous papers [8, 9]. The water content was less than 20 ppm. $LiAsF_6$ (Foot Minerals Co.), $LiPF_6$ (Hashimoto Kasei Co.) and $LiN(CF_3SO_2)_2$ (3M Corporation) were used as received. Hereafter, '1 M $LiAsF_6$–EC/PC(1:9)' will represent the electrolyte solution of the mixed solvents of EC and PC (mixing volume ratio = 1:9) in which 1 M (M:mol dm^{-3}) of $LiAsF_6$ is dissolved. Also, '[EC/PC]/2MeTHF ([85]: 15)' will represent the mixed solvents containing EC, PC and 2MeTHF where a total of 85 vol% of [EC + PC], and 15 vol% of 2MeTHF is mixed with PC.

2.2. *Fabrication of AA Li/a-V_2O_5–P_2O_5 cell*

The cell used was an AA-size laboratory-type cell with a structure very similar to that of the Li/MoS_2 cell fabricated by Moli Energy Ltd [10, 11]. This cell has a pressure vent in the bottom surface of the cell casing and is composed of a spirally wound Li anode sheet, a polyethylene separator, a printed cathode sheet of a-V_2O_5–P_2O_5 [12], polymer binder and conductive carbon. The cell used here had a capacity of about 600 mAh when the cell was cycled between 1.8 and 3.3 V, and an average discharge voltage of 2.55 V [2]. The energy density of the cell at 0.6 A discharge current was about 100 mAh g^{-1} and 200 mAh cm^{-3}.

The charge–discharge cycling tests for the AA-cells were carried out galvanostatically. The cell was operated at 21 °C unless otherwise noted. The cycle life of the AA-cells was evaluated by the figure of merit (FOM_{Li}) for Li cycling defined in Equation 1, by setting the end of life (EOL) at the cycle number where the discharge capacity becomes 50% of the maximum capacity of the cycles. The FOM_{Li} is related to the cycling efficiency of the lithium anode (E_{ff},%) as shown in Equation 2.

$$FOM_{Li} = [\text{accumulated discharge capacity}] / [\text{theoretical capacity of Li initially put into the cell}] \quad (1)$$

$$FOM_{Li} = 100/(100 - E_{ff}) \quad (2)$$

2.3. *Abuse test*

A heating test, in which the cells were placed in a preheated incubator, was employed as a fundamental abuse test.

3. Results and discussion

3.1. *EC/PC ratio in EC/PC/2MeTHF*

The influence of the mixing ratio of EC and PC in 1 M $LiAsF_6$–EC/PC/2MeTHF ternary mixed solvent systems on the cell properties of AA Li/a-V_2O_5–P_2O_5 cells was examined. In this investigation, the 2MeTHF content was fixed at 15 vol% as mentioned above. That is, the total amount of EC/PC in the ternary mixed solvents was 85 vol% and the four different EC/PC mixing ratios (5:80, 15:70, 30:55 and 42.5:42.5) were examined. These EC content values correspond to 6, 18, 35 and 50 vol%, respectively, when the total amount of EC and PC is assumed to be 100.

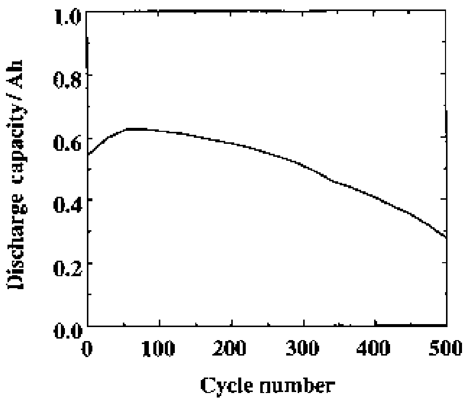

Fig. 1. Relationship between discharge capacity and cycle number of an AA size Li/a-V_2O_5–P_2O_5 cell containing 1 M $LiAsF_6$–EC/PC/2MeTHF (15:70:15) cycled between 1.8 V and 3.3 V with discharge and charge currents of 0.6 A and 0.1 A, respectively.

Figure 1 shows an example of the relationship between the discharge capacity and cycle number of an AA Li/a-V_2O_5–P_2O_5 cell containing 1 M $LiAsF_6$–EC/PC/2MeTHF (15:70:15) with discharge and charge currents of 0.6 A and 0.1 A, respectively, cycled between 1.8 V and 3.3 V. Figure 2 shows the relationship between the FOM_{Li} of the AA Li/a-V_2O_5–P_2O_5 cell and the EC content of EC/PC/2MeTHF. We used 3.3 V and 3.5 V as the charge cutoff voltages in this work. The FOM_{Li} of the cell at a charge cutoff of 3.3 V is higher than that at 3.5 V, while the capacity for charging at 3.5 V is larger than that at 3.3 V as mentioned later. With both the 3.3 V standard charge and the 3.5 V overcharge, the maximum FOM_{Li} of the AA cell is obtained at an EC/PC mixing ratio of 15:70 where EC content is 18% when the total EC/PC

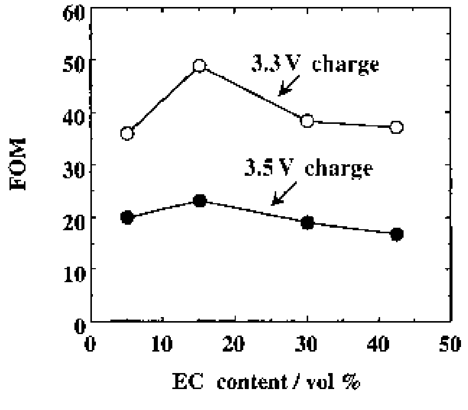

Fig. 2. Relationship between the FOM_{Li} of an AA Li/a-V_2O_5–P_2O_5 cell and the EC content in 1 M $LiAsF_6$–[EC/PC]/2MeTHF ([85]/15) mixed solvent electrolytes, with discharge and charge currents of 0.6 A and 0.1 A, respectively and a discharge cutoff voltage of 1.8 V.

amount is assumed to be 100. This result agrees with those of the previous studies [3] which reported that EC/PC binary mixed systems with small amounts of EC (10–30%) show the best FOM_{Li} for AA Li/a-V_2O_5–P_2O_5 cells.

Figure 3 shows the relationship between the electrolyte conductivity (κ) and the EC/PC mixing ratio. At 25 °C, the conductivity tends to increase gradually with an increase in EC content and to saturate around 30 vol% EC. This conductivity is 6.2 mS cm^{-1} and is just a little higher than the border line value of 4 mS cm^{-1} required for practical applications [13]. This low conductivity results from the high viscosity of EC/PC mixed with only 15 vol% 2MeTHF which is a low viscosity solvent. The viscosity of EC/PC/2MeTHF(15:70:15) measured here is 1.98 cP at 25 °C which is 82.5% of that for EC/PC. When a lithium salt with a large anion such as $LiAsF_6$ is used, solvent viscosity has a greater effect on the improvement in electrolyte conductivity than the dielectric constant because such a salt has a low reticular energy, and low migration speed [8]. Due to the large anions the reticular energy is low, so even with low dielectric constant, it is easily dissociated. The solvent viscosity must be less than 1 cP to realize a conductivity exceeding 10^{-2} S cm^{-1} at 25 °C based on our conductivity data for various electrolyte systems [8]. As the temperature is reduced, the EC content exhibiting the maximum conductivity tends to decrease. This is due to the higher activation energy obtained from the Arrhenius plot ($\log \kappa T$ vs the T plot, where T is the absolute temperature [14]), caused by the higher melting point of EC [15]. However, as shown in Fig. 3, in the −20–60 °C temperature range, the variation in conductivity caused by a change in EC content is very small and can be considered practically negligible.

Figure 4 shows the relationship between the capacity of an AA Li/a-V_2O_5–P_2O_5 cell and the EC content in EC/PC/2MeTHF. The AA cell capacity tends to reach maximum at an EC content of 15 vol%. However, the difference in the cell capacities is less than 8% in the EC content range of 5 to 42.5 vol% in EC/PC/2MeTHF. The EC/PC mixing ratio had no practical influence on capacity and this was because changes in the ratio made little difference to electrolyte conductivity.

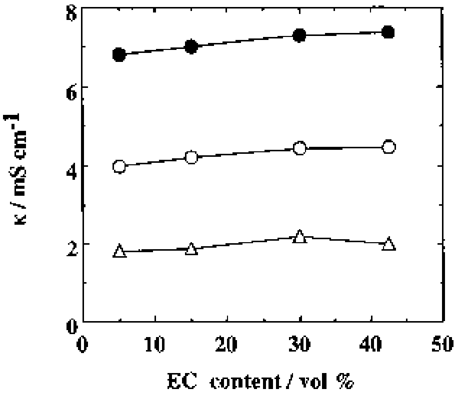

Fig. 3. Relationship between electrolyte conductivity (κ), temperature and EC content in 1 M $LiAsF_6$–[EC/PC]/2MeTHF ([85]/15) mixed solvent electrolytes. Temperature: (●) 25 °C, (○) −5 °C, (△) −15 °C.

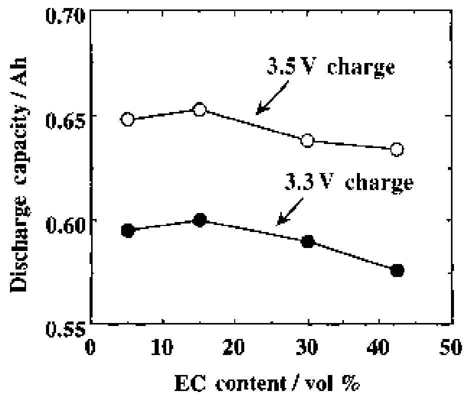

Fig. 4. Relationship between the capacity of an AA Li/a-V_2O_5–P_2O_5 cell and the EC content in 1 M $LiAsF_6$–[EC/PC]/2MeTHF ([85]/15) mixed solvent electrolytes, after cycling twice with discharge and charge currents of 0.6 A and 0.1 A, respectively and a discharge cutoff voltage of 1.8 V.

A heating test is a fundamental abuse test and is very effective for evaluating the safety of rechargeable lithium metal cells [16, 17]. An appropriate heating temperature is selected above the melting point of the separator, which may lead to an internal short between the anode and cathode. A temperature of 130 °C was adopted in this work because the cell has the polyethylene separator whose melting point is 125 °C. Heating test results at 130 °C for AA cells cycled using EC/PC/2MeTHF ternary mixed solvent electrolytes with different EC/PC mixing ratios indicate that the pressure vents of the cells remained closed and that the maximum cell skin temperature was between 135 and 145 °C. The cells were precycled before the heating tests at discharge and charge currents of 0.6 A and 0.1 A, respectively, between 3.3 V and 1.8 V (about 400 cycles) or between 3.5 V and 1.8 V (about 200 cycles) to the end of cycle. The heating test results obtained were substantially the same for all the AA cells examined. By contrast, when 130 °C heating tests were carried out for AA Li/a-V_2O_5–P_2O_5 cells, those with electrolytes containing 2MeTHF exhibited a higher maximum temperature than those with EC/PC binary mixed solvent electrolytes, as shown in Fig. 5. This larger heat output of electrolytes containing 2MeTHF resulted from an exothermic reaction between $LiAsF_6$ and 2MeTHF [7], that is, the decomposition of the organic electrolyte, as well as that between lithium and electrolyte [18, 19]. These results indicate that, in terms of safety, it is better to have a low 2MeTHF content.

Therefore, based on the overall results for cycle life, capacity and safety tests, the best solvent mixing composition for EC/PC/2MeTHF is 15:70:15 among the electrolytes examined here.

3.2. *Lithium salt concentration*

The influence of $LiAsF_6$ concentration on the FOM_{Li}, capacity and safety of the AA cells was ex-

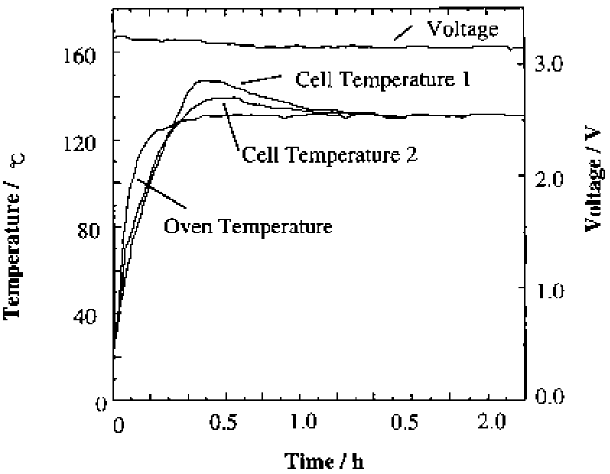

Fig. 5. Results of heating tests at 130 °C for AA Li/a-V_2O_5–P_2O_5 cells cycled 80 times at discharge and charge currents of 0.6 A and 0.1 A, respectively between 1.8 V and 3.5 V at 21 °C, cell temperature 1: 1 M $LiAsF_6$–EC/PC/2MeTHF (15:70:15), cell temperature 2: 1 M $LiAsF_6$–EC/PC (50:50).

amined. Four different $LiAsF_6$ concentrations (0.75, 1.00, 1.15 and 1.5 M) in EC/PC/2MeTHF (15:70:15) were used.

Figure 6 shows the relationship between electrolyte conductivity and $LiAsF_6$ concentration. The conductivity exhibits a maximum value with change in the $LiAsF_6$ concentration. With a decrease in the temperature, the $LiAsF_6$ concentration exhibiting the maximum conductivity tends to decrease. For example, the conductivity exhibited its maximum value for 1.15 M of $LiAsF_6$ at 40 °C, for 1.00 M of $LiAsF_6$ at 20 °C and for 0.75 M of $LiAsF_6$ at 0 °C. This is because of the increase in the degree of ionic association of the lithium salt based on the increase in the viscosity with a decrease in temperature [15, 20].

Figure 7 shows the relationship between the discharge capacity of an AA Li/a-V_2O_5–P_2O_5 cell and the $LiAsF_6$ concentration. The discharge capacity is obtained by two charge–discharge conditions of (i) a 0.6 A discharge and a 0.1 A charge between 1.8 V and 3.3 V at 21 °C and (ii) a 0.04 A discharge and a 0.08 A charge between 1.5 V and 3.3 V at −10 °C. The capacity reached a maximum at around 1.15 M of $LiAsF_6$ at a 0.6 A discharge at 21 °C and 1.5 M of $LiAsF_6$ exhibited the lowest value. This result is closely related to electrolyte conductivity. With a 0.04 A discharge at −10 °C, the capacity was similar for cells with different $LiAsF_6$ concentrations, except for 1.5 M $LiAsF_6$. 1.5 M $LiAsF_6$ exhibited the lowest capacity due to the low electrolyte conductivity.

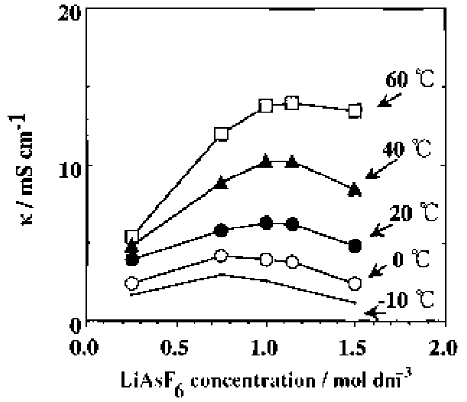

Fig. 6. Relationship between electrolyte conductivity (κ), temperature and $LiAsF_6$ concentration in EC/PC/2MeTHF (15:70:15).

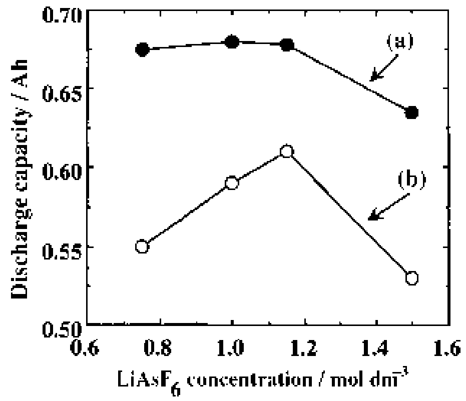

Fig. 7. Relationship between the capacity of AA Li/a-V_2O_5–P_2O_5 cells and $LiAsF_6$ concentration in EC/PC/2MeTHF (15:70:15) after cycling twice. (a) 0.6 A discharge and 0.1 A charge between 1.8 V and 3.3 V at 21 °C; (b) 0.04 A discharge and 0.08 A charge between 1.5 V and 3.3 V at −10 °C.

Figure 8 shows the relationship between the FOM_{Li} values of AA cells and the $LiAsF_6$ concentration. The cells were cycled under two different cycling conditions of (i) a 0.6 A discharge between 1.8 V and 3.3 V at 21 °C and (ii) a 0.04 A discharge between 1.5 V and 3.3 V at −10 °C. These charge–discharge conditions are the same as those used for the capacity evaluation described above. The FOM_{Li} increased for the cells cycled both at 21 °C and −10 °C with an increase in $LiAsF_6$ concentration. One reason why 1.5 M $LiAsF_6$ shows a larger FOM_{Li} than the others is the shallow discharge depth (low capacity), which is advantageous as regards improving the FOM_{Li} of both the anode and cathode [10, 19]. Another possible reason for the FOM_{Li} improvement with an increase in $LiAsF_6$ concentration may be

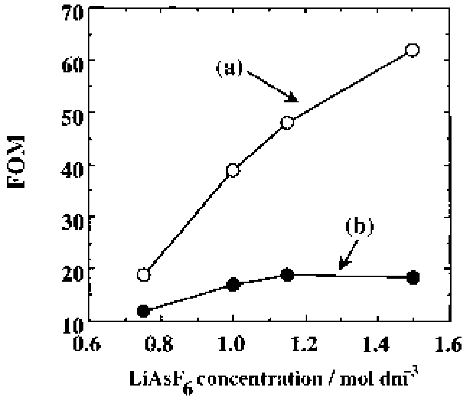

Fig. 8. Relationship between the FOM_{Li} of AA Li/a-V_2O_5–P_2O_5 cells and $LiAsF_6$ concentration in EC/PC/2MeTHF (15:70:15). (a) 0.6 A discharge and 0.1 A charge between 1.8 V and 3.3 V at 21 °C; (b) 0.04 A discharge and 0.08 A charge between 1.5 V and 3.3 V at −10 °C.

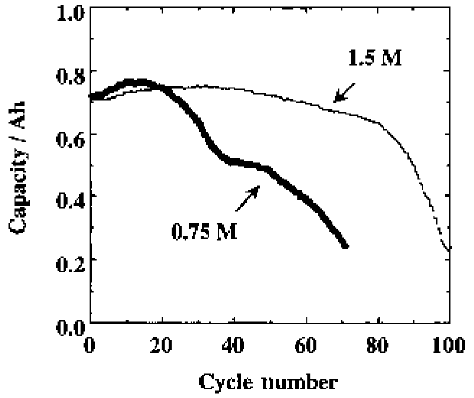

Fig. 9. Relationship between discharge capacity and cycle number for AA Li/a-V_2O_5–P_2O_5 cell with $LiAsF_6$–EC/PC/2MeTHF (15:70:15) cycled at discharge and charge currents of 0.06 A and 0.06 A, respectively, between 1.8 V and 3.3 V at 12 °C.

based on the formation of a suitable film on the Li surface which enhances the Li cycling [16, 19]. This film formation reaction involves the consumption of $LiAsF_6$ during each cycle [1, 6, 19, 21]. In addition, electrolyte depletion is reported as the reason for the end of the cycle life of lithium metal cells [19, 22]. This means that as cycle number increases, the amount of $LiAsF_6$ becomes smaller. For example, as shown in Fig. 9, 0.75 M $LiAsF_6$ showed a distinct two-step capacity fade curve, although 1.5 M $LiAsF_6$ does not show such a result. The second step may result from $LiAsF_6$ consumption, which causes high anode impedance (i) by the change in the surface film resulting from the reaction between Li and solvent rather than a $LiAsF_6$-related reaction and/or (ii) by the decrease in electrolyte conductivity.

The heating tests at 130 °C were carried out for AA Li/V_2O_5–P_2O_5 cells cycled at high rate discharge of 0.6 A using electrolytes with different $LiAsF_6$ concentrations. None of the cells showed any dangerous venting. However, the maximum cell temperature in the heating tests tended to decrease with increase in $LiAsF_6$ concentration (Fig. 10).

If the FOM_{Li} is the only concern, 1.5 M $LiAsF_6$ is the most suitable electrolyte concentration. However, 1.15 M $LiAsF_6$ is more appropriate in terms of overall performance including cyclability, capacity including low temperature behaviour, safety and cost from a practical point of view. This is because $LiAsF_6$ is the most expensive of the electrolyte materials.

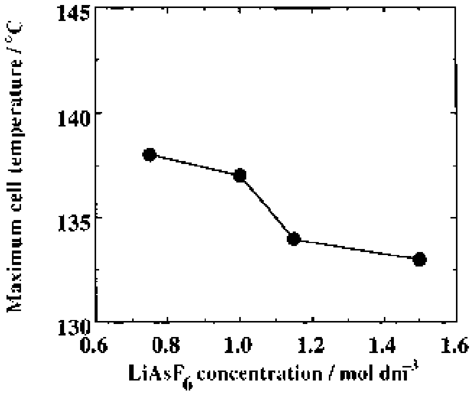

Fig. 10. Relationship between the maximum cell temperature of AA Li/a-V_2O_5–P_2O_5 cell by the heating test at 130 °C and $LiAsF_6$ concentration in EC/PC/2MeTHF (15:70:15), cycled at discharge and charge currents of 0.6 A and 0.1 A, respectively, between 1.8 V and 3.3 V at 21 °C.

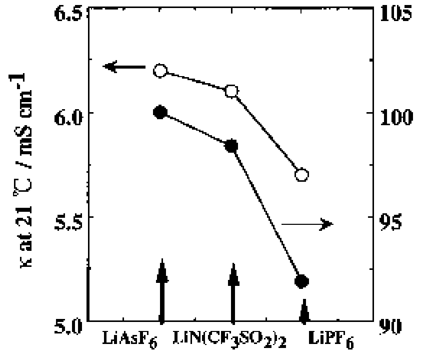

Fig. 11. Conductivity (κ) of 1 M lithium salt-EC/PC/2MeTHF (15:70:15) at 21 °C.

3.3. *Different lithium salts*

The influence of different lithium salts on the cell performance and safety of AA Li/a-V_2O_5–P_2O_5 cells was examined by comparing their properties with those of $LiAsF_6$. The lithium salts used here were $LiPF_6$ and $LiN(CF_3SO_2)_2$ which have a wide electrochemical window [1]. We used a salt concentration of 1 M and EC/PC/2MeTHF(15:70:15) as the solvent.

Figure 11 shows the conductivity (κ) of 1 M $LiPF_6$ or $LiN(CF_3SO_2)_2$–EC/PC/2MeTHF (15:70:15) compared with that of $LiAsF_6$ electrolyte. Figure 12 shows the relationship between conductivity and temperature. The conductivity of $LiPF_6$ is slightly lower than $LiAsF_6$ at 21 °C. This is because the degree of ionic dissociation of $LiPF_6$ is smaller than that of $LiAsF_6$ because PF_6^- (0.254 nm) has a slightly smaller anion radius than AsF_6^- (0.260 nm) [23]. The conductivity of $LiN(CF_3SO_2)_2$ is 8% lower than that of $LiAsF_6$ at 21 °C, because the contribution of anion migration to the conductivity is lower due to very large $N(CF_3SO_2)_2^-$ anion whose radius is 0.325 nm [23]. Below 0 °C, the conductivity values of these

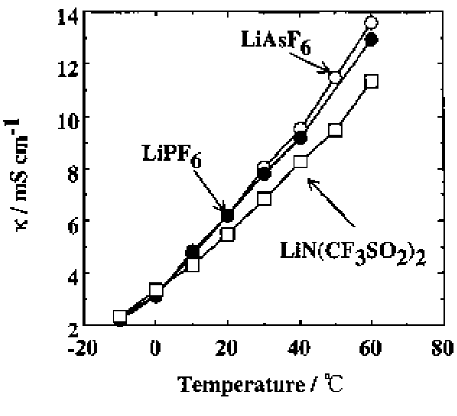

Fig. 12. Relationship between electrolyte conductivity (κ) and temperature for 1 M lithium salt-EC/PC/2MeTHF (15:70:15).

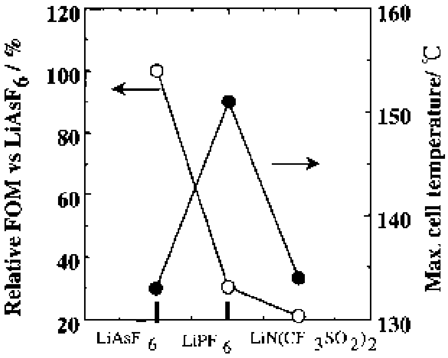

Fig. 13. The maximum cell temperature of AA Li/a-V_2O_5–P_2O_5 cells determined by a heating test at 130 °C and the relative FOM_{Li} vs $LiAsF_6$ for 1 M lithium salt–EC/PC/2MeTHF(15:70:15), cycled at discharge and charge currents of 0.6 A and 0.1 A, respectively, between 1.8 V and 3.3 V at 21 °C.

three salts become very similar. This is because of the high viscosity of EC/PC/2MeTHF as already mentioned.

Figure 13 shows the relative FOM_{Li} of AA cells with different lithium salts compared to that with $LiAsF_6$ electrolyte. The FOM_{Li} of an AA cell with $LiPF_6$ is similar to that with $LiN(CF_3SO_2)_2$, and are around 30% that with $LiAsF_6$. This result may relate to the decrease in lithium cycling efficiency caused by the formation of a surface film which is unsuitable for Li cycling because these three salts are reducible by Li [19] and $LiAsF_6$ is known to form a suitable film [1, 6, 19, 21]. Figure 13 also shows the maximum cell temperature determined by a heating test for AA cells cycled to the end of cycle. The maximum cell temperatures in the heating tests follow the order $LiAsF_6 < LiN(CF_3SO_2)_2 < LiPF_6$. This order is the same as that of the thermal stability of the salts themselves [24]. In addition, exothermic oxidation of 2MeTHF by $LiPF_6$ leading to extra heat output in the heating test could more easily occur than with the other salts because $LiPF_6$ is chemically and thermodynamically unstable [1, 18, 21, 24]. With $LiN(CF_3SO_2)_2$, the corrosion reaction of metal components such as the cell casing and electrode substrate, is possibly involved in the extra heat output [1].

In terms of both safety and cyclability, there is no reason to select $LiPF_6$ or $LiN(CF_3SO_2)_2$ salt rather than $LiAsF_6$ for the Li/a-V_2O_5–P_2O_5 cell system with EC/PC/2MeTHF solvents and with the cell configuration used here.

4. Conclusion

The influence of the composition of EC/PC/2MeTHF (2MeTHF:15 vol%) ternary mixed solvent electrolyte on the charge–discharge properties of AA Li/a-V_2O_5–P_2O_5 cells was examined. A heating test as a fundamental abuse test on the cells was also performed. The results revealed the following: (i) with regard to the appropriate EC/PC mixing ratio, EC/PC (15:70) showed the highest capacity and the longest cycle life; (ii) with regard to $LiAsF_6$ concentration, 1.15 M is the most suitable based on the overall cycle life and capacity including low temperature behaviour; and (iii) with regard to the kind of lithium salt to be used, $LiAsF_6$ exhibits a much longer cycle life than $LiPF_6$ or $LiN(CF_3SO_2)_2$.

On the basis of cell performance, cost and safety test results, the best composition among the electrolytes for AA Li/a-V_2O_5–P_2O_5 cell is 1.15 M $LiAsF_6$–EC/PC/2MeTHF (15:70:15).

References

[1] G. Pistoia (ed.), 'Industrial Chemistry Library', vol.5, Lithium Batteries, New Materials, Developments and Perspectives, Elsevier, Netherlands (1994), chap. 4.
[2] Y. Sakurai, S. Sugihara, M. Shibata and J. Yamaki, *NTT Rev.*, **7** (1995) 60.
[3] S. Tobishima, K. Hayashi, K. Saito, T. Shodai and J. Yamaki, *Electrochim. Acta*, **42** (1997) 119.
[4] M. Arakawa, Y. Nemoto, S. Tobishima and M. Ichimura, *J. Power Sources* **43/44** (1993) 517.
[5] S. Tobishima, K. Hayashi, Y. Nemoto and J. Yamaki, *Electrochim. Acta*, in press.
[6] 'Catalog Handbook of Fine Chemicals 1994–1995', Aldrich Chemical Company, Inc.
[7] M. A. Gee and F. C. Laman, *J. Electrochem. Soc.* **140** (1993) L53.
[8] S. Tobishima and A. Yamaji, *Electrochim. Acta* **29** (1984) 267.
[9] S. Tobishima and T. Okada, *ibid.* **30** (1985) 1715.
[10] F. C. Laman and K. Brandt, *J. Power Sources* **21** (1987) 195.
[11] MOLICEL product catalogue, '1:Introduction to MOLICEL, Cells and Batteries', Moli Energy Ltd, Canada (1987).
[12] Y. Sakurai and J. Yamaki, *J. Electrochem. Soc.* **132** (1985) 512.
[13] J. P. Gabano (Ed.), 'Lithium Batteries', Academic Press, (1983), chapters 2 and 13.
[14] I. D. Raistrick, C. Ho and R. A. Huggins, *Mat. Res. Bull.* **11** (1976) 953.
[15] S. Tobishima, K. Hayashi, K. Saito and J. Yamaki, *Electrochim. Acta* **40** (1995) 537.
[16] D. P. Wilkinson and J. Dahn, Extended Abstracts of Electrochemical Society Fall Meeting, Abstract 53, Seattle, Washington (1990), p. 85.
[17] L. Lechmer and H. Woo, Extended Abstracts of Electrochemical Society Fall Meeting, Abstract 14, Phoenix, Arizona (1991), p. 20.
[18] V. R. Koch, *J. Power Sources* **6** (1981) 357.
[19] K. M. Abraham, *Electrochim. Acta.* **38** (1993) 1233.
[20] K. M. Abraham and J. L. Goldman, *J. Power Sources* **9** (1983) 239.
[21] V. R. Koch, *J. Electrochem. Soc.* **126** (1979) 181.
[22] K. M. Abraham, J. S. Foos and J. L. Goldman, *J. Electrochem. Soc.* **131** (1984) 2197.
[23] M. Ue, *ibid.* **141** (1994) 3336.
[24] J. T. Dudley, D. P. Wilkinson, G. Thomas, R. Levae, S. Woo, H. Blom, C. Horvan, M. W. Juzkow, B. Denis, P. Juric, P. Aghakian and J. R. Dahn, *J. Power Sources* **35** (1991) 59.