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A single-ion gel polymer electrolyte based on polymeric lithium tartaric acid borate and its superior battery performance

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ABSTRACT

A single-ion gel polymer electrolyte of PLTB@PVDF-HFP (polymeric lithium tartaric acid borate@poly(vinylidene fluoride-co-hexafluoropropene)) was prepared and its ionic conductivity was optimized via solvent composition tailoring. It was manifested that the ethylene carbonate/dimethyl carbonate (EC/DMC) swollen PLTB@PVDF-HFP exhibited a superior lithium ionic conductivity at room temperature to that of the traditional liquid electrolyte system (1 M LiPF₆/EC/DMC). The Li₄Ti₅O₁₂/LiFePO₄ cells using the EC/DMC swollen PLTB@PVDF-HFP as polymer electrolyte showed stable charge/discharge voltage profiles with small voltage hysteresis, preferable rate capability and excellent cycle performance at room temperature. These superior performances of EC/DMC swollen PLTB@PVDF-HFP could endow this class of gel polymer electrolyte a very promising alternative to state of the art liquid electrolyte system.

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1. Introduction

Lithium ion batteries have been attracted much attention in the last decade for their potential application for electric vehicles (EVs), hybrid electric vehicles (HEVs) and smart grid [1,2]. However, the large-capacity battery modulus claims more safety concerns, because fire or explosion at abnormal conditions may lead to serious hazards owing to the hidden peril of traditional liquid electrolyte system, i.e., lithium hexafluorophosphate solutions in aprotic organic solvents. This traditional liquid electrolyte system has been widely used in state of the art technology mainly owing to its high ionic conductivity (10^{-3} – 10^{-2} S cm⁻¹) at room temperature [3–9]. The use of a gel polymer electrolyte (GPE) in place of the liquid electrolyte may help to tackle safety concerns: (i) suppression of lithium dendrite growth that is usually caused by uneven currents when charged in the case of porous separators. (ii) Alleviation of interfacial reactivity between the electrode and liquid electrolytes due to the least use of alkyl carbonate solvents. (iii) Improvement of safety, in addition to suppression of lithium dendrites, the quasi-solid-state construction of a polymer electrolyte is more tolerant to mechanical deformation. (iv) Better shape flexibility and manufacturing integrity [10–17]. Meanwhile, to be successfully used in practice, GPE must meet the following basic requirements: (i) ionic conductivity higher than $\sim 10^{-4}$ S cm⁻¹ at room temperature, (ii) lithium ion transference number close to unity, (iii) good thermal, chemical and electrochemical stability and (iv) a considerable mechanical strength [10,12,18–21]. However, rare GPEs have been reported to accomplish all above standards.

Lithium hexafluorophosphate (LiPF₆) is predominant lithium salt in liquid electrolyte system [9,10]. However, LiPF₆ is known to be extremely sensitive to moisture and thermally decompose at about 60 °C. Its decomposition components consist of high toxic and corrosive HF, PF₅ and POF₃, where HF can dissolve cathode active materials, PF₅ can react with electrolyte solvents and consequently bring about thermal runaway [22–24]. In the last few years, a kind of lithium borate salts, for example, lithium bis(oxalato) borate (LiBOB) has attracted great interest to researchers because of superior thermal stability, considerable ionic conductivity and fair dissociation constant [25–27]. Furthermore, some kinds of polymer electrolytes based on chelate compound with boron were also reported. Angell et al. have developed polymer electrolytes based on hybrid composition of LiBOB and poly(ethylene glycol)s (PEGs) with the ionic conductivities reaching $\sim 10^{-5}$ S cm⁻¹ at room temperature. And the lithium ion transference number was far away from the theoretical value even for the polyanionic solid electrolytes, owing to the presence of some anion impurity produced unavoidably during synthesis [28–30]. Two kinds of single-ion polymer electrolytes, lithium oxalate polyacrylic acid borate (LiPAAOB) and lithium polyvinyl alcohol oxalate borate (LiPVAOB) were very recently synthesized by Wu Y. P. et al. [31,32]. While, the ionic conductivities for both of them could only reach $\sim 10^{-6}$ S cm⁻¹ at room temperature, far away from the requirement for applicable polymer electrolytes for rechargeable lithium ion batteries.

In our previous report, a novel kind of “main-chain” type polymeric lithium tartaric acid borate (PLTB, Fig. 1(a)) was synthesized via a one-step reaction in aqueous solution [33]. The as-prepared PLTB possesses a high thermal decomposition temperature at 330 °C. The GPE of PLTB@PVDF-HFP was developed by a doctor-blading followed by a soaking process in propylene carbonate (PC). It was manifested that the PC

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Fig. 1. (a) Chemical structure of the polymeric lithium tartaric acid borate (PLTB), (b) photograph of the PLTB@PVDF-HFP membrane and (c) its surface morphology observed by SEM.

swollen PLTB@PVDF-HFP exhibited excellent properties at an operating temperature of 80 °C. However, PC swollen PLTB@PVDF-HFP suffered poor rate capability at room temperature, which would limit its extensive application because most of battery devices are indeed operated at ambient temperature. In this paper, we optimized the ion conductivity of PLTB@PVDF-HFP membranes in mixed solvents instead of PC to further improve the ionic conductivity at room temperature. The battery properties including charge/discharge behavior, rate capability and cycling performance of mixed solvent swollen PLTB@PVDF-HFP were investigated in $\text{Li}_4\text{Ti}_5\text{O}_{12}/\text{LiFePO}_4$ full cells at room temperature. Since most of the irreversible reactions are thermally activated by the rise in temperature in an Arrhenius-like manner, their battery properties were also compared with those of the traditional liquid electrolyte cells in an accelerated manner at high temperature of 60 °C.

2. Experimental

2.1. Materials

Ethylene carbonate (EC), propylene carbonate (PC) and dimethyl carbonate (DMC) (all from Capchem Technology Co., Ltd), 1 M LiPF_6 -EC/DMC (1/1, v/v, Guotai-huarong New Chemical Materials Co., Ltd.), PVDF-HFP (Aldrich, $M_w \sim 400,000$, $M_n \sim 130,000$, pellets, VDF:HFP $\sim 1:1$), N,N-Dimethyl Formamide (99.8%, superdry, J&K), carbon-coated LiFePO_4 (Tianjin STL Energy Technology Co., Ltd.), carbon-coated $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (BTR Nano Technology Co., Ltd.), PP separator (Celgard 2500). All materials were all commercially available and used without further purification.

2.2. Preparation of mixed solvent swollen PLTB@PVDF-HFP membrane

The procedure for the synthesis of PLTB was presented in our previous work [33]. The preparation of PLTB@PVDF-HFP membranes are described briefly as follows. PLTB and PVDF-HFP were added into DMF (N,N-dimethyl formamide) and mixed to be a transparent and homogeneous solution. Then the PLTB@PVDF-HFP membranes were prepared by a doctor-blading process under ambient atmosphere

followed by a vacuum drying at 80 °C to remove DMF. Finally, PLTB@PVDF-HFP membranes (PLTB/PVDF-HFP = 1.0/1.0 (w/w), 25 μm) were immersed in mixed solvents to obtain the solvent swollen PLTB@PVDF-HFP membrane. The thickness of solvent swollen PLTB@PVDF-HFP membrane was $\sim 75 \mu\text{m}$.

2.3. Characterization of mixed solvent swollen PLTB@PVDF-HFP membrane

The surface morphology of PLTB@PVDF-HFP membrane was observed by Hitachi S-4800 field emission scanning electron microscope. The mechanical property of the membrane was measured using an Inston-3300 universal testing machine (USA) at a stretching speed of 1.66 mm/s with the straps of about 1 cm wide and 5 cm long.

The ionic conductivity of solvent swollen PLTB@PVDF-HFP membrane between two stainless-steel plate electrodes was obtained by an AC impedance analysis using a Zahner Zennium electrochemical working station over a frequency range of 1–10⁶ Hz with a perturbation amplitude of 10 mV.

The electrochemical stability of solvent swollen PLTB@PVDF-HFP membrane was evaluated by a cyclic voltammetry experiment performed on a working electrode of stainless-steel and a counter electrode of lithium metal at a scan rate of 0.1 mV/s. Lithium ion transference number was measured according to the method described by Evans et al. [34]. The method involved sandwiching the sample membrane in both lithium electrodes. After having measured the total initial resistance by AC impedance, a DC potential of 10 mV was applied until a steady state was reached. Finally, the resistance was again measured by AC impedance.

$$t^+ = \frac{I_s(\Delta V - I_0 R_0^{el})}{I_0(\Delta V - I_s R_s^{el})}$$

where t^+ is the cationic transference number, ΔV is the potential applied across the cell, R_0^{el} and R_s^{el} are the initial and steady-state resistances of the passivating layers on the Li electrode and I_0 and I_s are the initial and steady-state currents.

2.4. Cell assembly and performance

The Li/PLTB@PVDF-HFP/Li₄Ti₅O₁₂ half coin cells (2032-type) were assembled by sandwiching the EC/DMC (1/1, v/v) swollen PLTB@PVDF-HFP membrane between lithium metal foil and Li₄Ti₅O₁₂ electrode. The Li₄Ti₅O₁₂/PLTB@PVDF-HFP/LiFePO₄ full coin cells (2032-type) were also assembled with LiFePO₄ instead of lithium metal foil. The Li₄Ti₅O₁₂ electrode was composed of 85 wt.% Li₄Ti₅O₁₂ (around 2.7 mg active material on 1.54 cm² copper metal foil), 7.5 wt.% PVDF and 7.5 wt.% carbon black. The LiFePO₄ electrode was composed of 85 wt.% LiFePO₄ (around 3.0 mg active material on 1.54 cm² aluminum metal foil), 7.5 wt.% PVDF and 7.5 wt.% carbon black. All assembly of cells was carried out in an argon-filled glove box. After cell assembly, the cells were laid down on one side for 12 h and then on the other side for another 12 h to impregnate the porous composite electrodes and their interfaces with electrolyte. The charge/discharge, C-rate capacity and cycling ability of cells were recorded on a LAND battery test system. The galvanostatic charge/discharge behavior of Li/PLTB@PVDF-HFP/Li₄Ti₅O₁₂ cells and Li₄Ti₅O₁₂/PLTB@PVDF-HFP/LiFePO₄ cells were conducted over the range of 1.0–3.0 V and 1.0–2.7 V, respectively.

3. Results and discussion

3.1. Characterization of PLTB@PVDF-HFP membrane

The surface morphology of PLTB@PVDF-HFP membrane observed by SEM was depicted in Fig. 1. It could be seen that the PLTB@PVDF-HFP membrane was quite homogeneous and there was no obvious pores on the surface. The membrane exhibited tensile strength of 19 MPa with 53% strain (seen in Fig. S1), which indicated that the polymer electrolyte membranes possessed a considerable mechanical strength. After swollen with solvents, the gel electrolyte possessed tensile strength of 5.0 MPa with 133% strain.

3.2. Ionic conductivity optimization of PLTB@PVDF-HFP swollen in mixed solvent

It is well known that gel polymer electrolytes usually have an inferior ionic conductivity when compared with traditional liquid electrolytes, which impairs the power performance of gel polymer batteries [9,10,18]. Three strategies are consequently involved to improve the power performance. The first one is to tailor solvent composition for manipulating electrolyte ion conductivity due to the availability of a vast number of candidate solvents [13,35]. The most important are two parameters of the solvents, dielectric constant (ϵ) and viscosity (η), which determine the charge carrier number and ion mobility, respectively. The second one is to enhance the ionic conductivity of gel polymer electrolytes by compositing with nanoparticles including SiO₂, lithium titanium phosphate, etc. [10,36]. The third one is to introduce single ionic conductor into the polymer matrices to improve the lithium ion transference number [31–33]. The as-prepared PLTB belongs to a single-ion conductor and the lithium ion transference number of PLTB@PVDF-HFP membranes can reach the theoretical value. Herein, we would like to exploit the first strategy to further improve the ionic conductivity of PLTB@PVDF-HFP membranes at room temperature through solvent composition tailoring.

Fig. 2 showed the temperature dependence of ionic conductivity for PLTB@PVDF-HFP membrane swollen in various solvents, such as PC, EC/PC (1/1, v/v) and EC/DMC (1/1, v/v). The mass ratio of PLTB/PVDF-HFP/solvent was 0.5/0.5/2.0 (w/w/w). It could be seen from Fig. 2 that the membranes swollen in mixed solvent showed higher ionic conductivity as compared with the membrane swollen in pure PC solvent. The reason for the improvement of ionic conductivity in mixed solvent is related to the more rapid decrease in viscosity than the dielectric constant when the low viscosity solvent is mixed with the high dielectric constant solvent [10,13]. The ionic conductivities

Fig. 2. Temperature dependence of ionic conductivity for PLTB@PVDF-HFP membranes swollen in various solvents. (PLTB/PVDF-HFP/solvent = 0.5/0.5/2.0, w/w/w.)

were 0.26 mS cm⁻¹, 0.35 mS cm⁻¹ and 0.50 mS cm⁻¹ at room temperature for PC, EC/PC and EC/DMC, respectively. PLTB@PVDF-HFP membrane soaked in EC/DMC possessed the optimal ionic conductivity over the range of 10 °C to 80 °C, so we chose EC/DMC soaked PLTB@PVDF-HFP membranes for further research in our experiments.

For traditional liquid electrolytes incorporated with polyolefin separators, the apparent ionic conductivities at room temperature generally kept at 0.1–1 mS cm⁻¹ and the lithium ion transference numbers ranged from 0.2 to 0.4, depending on the properties of salts and solvents (for example, 1 M LiPF₆-EC/DMC using Celgard 2500 as separator, $\sigma = 0.85$ mS cm⁻¹, $t_{\text{Li}^+} = 0.32$) [10]. A lithium ion transference number significantly less than 1.0 is certainly undesired, because the resultant overwhelming anion movement and enrichment near electrode surfaces would cause concentration polarization during battery operation. In addition, taking lithium ion transference number into consideration, the solvent swollen PLTB@PVDF-HFP delivered much higher lithium ionic conductivity (0.455 mS cm⁻¹) than that of traditional liquid electrolytes (0.27 mS cm⁻¹). Lithium ionic conductivity comparison for both electrolyte systems was listed in Table 1. The results indicated that this kind of gel polymer electrolyte could endow superior battery performance.

3.3. Electrochemical stability and Li⁺ transference number of EC/DMC swollen PLTB@PVDF-HFP membrane

The electrochemical stability of EC/DMC swollen PLTB@PVDF-HFP membrane was measured by the cyclic voltammetry. It could be seen from Fig. 3(a) that the EC/DMC swollen PLTB@PVDF-HFP membrane didn't oxidatively decompose until 5.0 V vs. Li⁺/Li at 20 °C. Also, the EC/DMC swollen PLTB@PVDF-HFP membrane showed very excellent lithium deposition-stripping performance. The transference number of Li⁺ was measured and calculated to be 0.91 using the steady-state current method (see Fig. 3(b)). This remarkably high transference number was attributed to the polyanion structure of the PLTB.

3.4. Battery performance of Li/PLTB@PVDF-HFP/Li₄Ti₅O₁₂ half cell

As a promising anode material for lithium ion batteries, spinel Li₄Ti₅O₁₂ exhibits many advantages compared to the currently used

Table 1
Apparent ionic conductivity comparison for both electrolyte systems.

1 M LiPF ₆ -EC/DMC (with separator)			EC/DMC swollen PLTB@PVDF-HFP		
σ /mS cm ⁻¹	t_{Li^+}	$\sigma_{\text{Li}^+}^a$ /mS cm ⁻¹	σ /mS cm ⁻¹	t_{Li^+}	$\sigma_{\text{Li}^+}^a$ /mS cm ⁻¹
0.85	0.32	0.27	0.50	0.91	0.455

^a $\sigma_{\text{Li}^+} = \sigma \times t_{\text{Li}^+}$.

Fig. 3. (a) Electrochemical stability of EC/DMC swollen PLTB@PVDF-HFP membrane at 20 °C (scan rate = 0.1 mV s⁻¹) and (b) variation of current with time during polarization of a Li/EC/DMC swollen PLTB@PVDF-HFP membrane//Li cell at 20 °C, with total applied potential difference of 10 mV.

graphite. For example, there is negligible volume change in the unit cell volume of Li₄Ti₅O₁₂ during lithium insertion and extraction, so it was named as zero strain material. It also has a high voltage plateau at around 1.55 V vs. Li⁺/Li. Therefore, Li₄Ti₅O₁₂ is much safer and more stable than carbon-based materials. In spite of gasing problem, the carbon-coated Li₄Ti₅O₁₂ was reported to possess good cycling performance. Fig. 4(a) showed the first charge/discharge voltage profiles of Li/EC/DMC swollen PLTB@PVDF-HFP//Li₄Ti₅O₁₂ at the temperature of 20 °C and 60 °C, and a stable voltage plateau at around 1.55 V vs. Li⁺/Li could be observed for both of them. The cycling performances of Li/Li₄Ti₅O₁₂ half cells using EC/DMC swollen PLTB@PVDF-HFP and 1 M LiPF₆-EC/DMC were shown in Fig. 4(b), where the charge/discharge rates were fixed at 0.5 C. The discharge capacity of the cells could retain 142.2 mAh g⁻¹ (calculated on the mass of Li₄Ti₅O₁₂) and 138.7 mAh g⁻¹ for LiPF₆-based electrolyte and EC/DMC swollen PLTB@PVDF-HFP after 60 cycles at 20 °C, respectively. The capacity retention, $R_{60/1}$, defined as ratio of capacity at 60th cycle over that at first cycle, was 98.1% and 92.8%, respectively. However, the discharge capacity of gel PLTB@PVDF-HFP cells could retain 146.4 mAh g⁻¹ after 60 cycles and $R_{60/1}$ was 99.0% at high temperature of 60 °C. It could be seen that the Li/Li₄Ti₅O₁₂ cells using EC/DMC swollen PLTB@PVDF-HFP membranes were comparable to those using LiPF₆ based liquid electrolytes.

3.5. Battery performance of Li₄Ti₅O₁₂/PLTB@PVDF-HFP/LiFePO₄ cell

The initial charge/discharge performances of Li₄Ti₅O₁₂/EC/DMC swollen PLTB@PVDF-HFP/LiFePO₄ cells at 20 °C and 60 °C were

Fig. 4. (a) The first charge/discharge voltage profiles of Li/EC/DMC swollen PLTB@PVDF-HFP//Li₄Ti₅O₁₂ cell and (b) cycling stability of Li/EC/DMC swollen PLTB@PVDF-HFP//Li₄Ti₅O₁₂ cells and Li/LiPF₆-EC/DMC//Li₄Ti₅O₁₂ cell at 20 °C or 60 °C. Rate: 0.5 °C.

presented in Fig. 5(a) and (b), respectively. Both Li₄Ti₅O₁₂/EC/DMC swollen PLTB@PVDF-HFP/LiFePO₄ cells had stable charge/discharge plateaus at 20 °C and 60 °C, which meant that PLTB@PVDF-HFP based polymer electrolyte had good interface compatibility with Li₄Ti₅O₁₂ and LiFePO₄ electrodes. The polarization and resultant voltage difference of PLTB@PVDF-HFP based cells was comparable to that of LiPF₆ electrolyte based cells under 0.5 C at ambient temperature. This was absolutely attributed to the excellent lithium ion conductivity of PLTB@PVDF-HFP electrolyte. While, at the elevated temperature of 60 °C, the Li₄Ti₅O₁₂/LiFePO₄ cell using LiPF₆-EC/DMC electrolyte failed to be initially charged and discharged smoothly. The discharge voltage profiles of PLTB@PVDF-HFP based cells at different discharge rates were depicted in Fig. 5(c) under 20 °C. It could be seen that the discharge capacities of the cells were 134.6 mAh g⁻¹ (based on the mass of LiFePO₄ and the geometric current density 25.5 μA/cm²), 132.9 mAh g⁻¹ (51 μA/cm²), 123.6 mAh g⁻¹ (127 μA/cm²) and 104.9 mAh g⁻¹ (255 μA/cm²), respectively. These results demonstrated that the Li₄Ti₅O₁₂/EC/DMC swollen PLTB@PVDF-HFP/LiFePO₄ cells had a considerable rate performance even at room temperature.

Fig. 6 showed the cycling performances of Li₄Ti₅O₁₂/LiFePO₄ cells using EC/DMC swollen PLTB@PVDF-HFP as electrolyte with the charge/discharge rates fixed at 0.5 C. It could be seen from Fig. 6(a) and (c) that the capacities of the cells using the polymer electrolyte retained 116.5 mAh g⁻¹ at 20 °C and 101.6 mAh g⁻¹ at 60 °C after 100 cycles. In the capacity retention, $R_{100/1}$ was 86.6% and 86.3% respectively. For LiPF₆ based cell at 20 °C, the discharge capacity was 110.3 mAh g⁻¹ at

Fig. 5. The first charge/discharge curves of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ //EC/DMC swollen PLTB@PVDF-HFP//LiFePO₄ cells at (a) 20 °C and (b) 60 °C, rate: 0.5 C. (c) Discharge voltage profiles of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ //EC/DMC swollen PLTB@PVDF-HFP//LiFePO₄ cells at different discharge rates. Temperature: 20 °C.

the 100th cycle and the discharge capacity retention was about 81.6%. The initial coulombic efficiencies were very similar for both PLTB@PVDF-HFP based cells (84.3%) and LiPF₆ based cells (85.6%) at room temperature (Fig. 6(b)). At the temperature of 60 °C, the coulombic efficiency of the first cycle for PLTB@PVDF-HFP based cell was 80.4% (Fig. 6(c)). All the other coulombic efficiencies of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ //EC/DMC swollen PLTB@PVDF-HFP//LiFePO₄ cells were nearly approaching to 100% in the following cycles, which could be seen from Fig. 6(b) and (c). However, both the capacities and the coulombic efficiencies of LiPF₆ based cell showed

Fig. 6. (a) Cycling stability and (b) coulombic efficiencies of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ //EC/DMC swollen PLTB@PVDF-HFP//LiFePO₄ cell and $\text{Li}_4\text{Ti}_5\text{O}_{12}$ //LiPF₆-EC/DMC//LiFePO₄ cell at 20 °C, (c) cycling stability and coulombic efficiencies of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ //EC/DMC swollen PLTB@PVDF-HFP//LiFePO₄ cell at 60 °C.

accelerated fading from about the 60th cycle at 20 °C. This implied that the SEI layer in PLTB@PVDF-HFP based cells was more stable and compatible than that in LiPF₆ based cells. Besides, the SEI layer exhibited a good thermal stability. Therefore, it was demonstrated that the cycling performance of PLTB@PVDF-HFP based cell were much enhanced than those of LiPF₆ based cell.

To further investigate the superior cycling performance of EC/DMC swollen PLTB@PVDF-HFP in $\text{Li}_4\text{Ti}_5\text{O}_{12}$ //LiFePO₄ cells, the surface morphology of LiFePO₄ electrode was studied by SEM after long battery

Fig. 7. SEM images of (a) pristine LiFePO₄ electrode, (b) the LiFePO₄ electrode from Li₄Ti₅O₁₂//EC/DMC swollen PLTB@PVDF-HFP//LiFePO₄ cell after 100 cycles at 60 °C and (c) the LiFePO₄ electrode from Li₄Ti₅O₁₂//LiPF₆-EC/DMC//LiFePO₄ cell after 100 cycles at 50 °C.

cycles at 0.5 C, which was depicted in Fig. 7. For comparison, the surface morphology of LiFePO₄ electrodes after cycling using LiPF₆-EC/DMC as electrolyte at 50 °C was also provided. It could be observed that some of the LiFePO₄ particles in the LiPF₆ based cell were apparently corroded and cracked after 100 cycles (Fig. 7(c)). While, the surface morphology of LiFePO₄ electrode in PLTB@PVDF-HFP based cell after 100 cycles at 60 °C (Fig. 7(b)) were almost the same as that of the pristine LiFePO₄ electrode (Fig. 7(a)). This implied that PLTB@PVDF-HFP based electrolyte was quite stable and compatible with LiFePO₄ electrode without noticeable corrosion after a long cycling time. The main reason for the slight decrease of discharge capacity after the repeated cycles was from the occurrence of the irreversible phase and structure within active material LiFePO₄, not related to the PLTB@PVDF-HFP electrolyte [37,38].

4. Conclusions

In summary, a high performance single-ion polymer electrolyte based on EC/DMC swollen PLTB@PVDF-HFP was investigated and compared with that of LiPF₆ based electrolyte. The EC/DMC swollen PLTB@PVDF-HFP exhibits a superior lithium ionic conductivity at room temperature ($\sigma_{\text{Li}^+} = 0.45 \text{ mS cm}^{-1}$) and significantly high lithium ion transference number ($t_{\text{Li}^+} = 0.91$). The Li₄Ti₅O₁₂/LiFePO₄ full cells using EC/DMC swollen PLTB@PVDF-HFP membranes exhibit stable charge/discharge plateau with small voltage difference, satisfactory rate performance and excellent cycling performance at room temperature. These superior performances of EC/DMC swollen PLTB@PVDF-HFP could endow this class of gel polymer electrolyte an excellent candidate electrolyte for high power lithium ion batteries.

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Appendix A. Supplementary data

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