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Journal Name

ARTICLE

One-step radiation synthesis of gel polymer electrolyte with high ionic conductivity for lithium-ion batteries

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We demonstrated a one-step synthesis method for a novel gel polymer electrolyte (named as PDMP-Li GPE) based on 3-(dimethylamino) propyl methacrylate, poly(ethylene glycol) diacrylate and 1 M LiPF₆ liquid electrolyte solution by γ -radiation technique for lithium-ion battery. The resultant PDMP-Li GPE exhibited good thermal stability, excellent mechanical strength and high ionic conductivity, and the highest ionic conductivity reached $8.88 \times 10^{-3} \text{ S cm}^{-1}$ at 25 °C. The LiFePO₄/PDMP-Li GPE/graphite pouch-type cell exhibited more than 97% coulombic efficiency and the capacity retention was about 86% after 50 cycles at 0.1 C. With the outstanding characteristics, it is expected to be a potential candidate for the application in polymer lithium-ion batteries.

Introduction

Lithium-ion batteries have gained an unprecedented importance in the last several decades as the energy storage of portable devices such as cell phones, laptop computers and so on.¹⁻⁴ However, some serious problems including leakage, flame and explosion have been reported to happen in the application of lithium-ion batteries with liquid electrolytes, because of the existence of highly flammable organic liquid electrolytes.⁵ The solvent-free solid electrolyte is the ideal way to solve the current safety problem of liquid electrolyte, but this advantage goes along with a considerably reduced conductivity.⁶ To combine the positive properties of liquid and solid polymer electrolytes, gel polymer electrolytes (GPEs) have been introduced, which have higher ionic conductivity and more stability than solid polymer electrolytes and liquid electrolytes, respectively, and consequently they could provide an alternative solution to safety problem of lithium-ion battery.^{7,8} Furthermore, compared with the commercial liquid electrolytes, the main advantages of GPEs in lithium-ion batteries are its non-leakage nature, higher thermal stability, and it can be used to prepare the flexible, thinner and smaller electrochemical devices.

Recently, various polymers such as poly(acrylonitrile) (PAN)^{9,10}, poly(vinylidene fluoride) (PVDF)^{11,12} and poly(methyl methacrylate) (PMMA)¹³⁻¹⁵ have been widely used as polymer matrices for the preparation of GPEs in the lithium-ion batteries. The ionic conductivity of these systems was around 10^{-5} - $10^{-3} \text{ S cm}^{-1}$ at room temperature, which was influenced by

the uptake amount of the liquid electrolyte within the polymer matrix.¹⁶ However, the increase of liquid electrolyte uptake brings the low mechanical properties of GPEs, that is, the conductivity and mechanical stability of GPEs are mutually exclusive. Notably, PMMA has received considerable attention due to its similar structure to carbonate-based solvents applied in the conventional liquid electrolytes, which suggests that it may have good compatibility with the lithium salts as well as offer a good interfacial contact with commonly used electrodes. Meanwhile, its amorphous structure is beneficial to the ionic conductivity ($1 \times 10^{-3} \text{ S cm}^{-1}$ at ambient temperature), but still can not meet the needs of practical applications.

Besides, the GPEs can be prepared by ultraviolet-curing (UV)^{17,18}, solvent-casting technique^{19,20}, thermal method^{16,21} and electrospinning technique²²⁻²⁵. Compared with traditional methods, γ -radiation is a simpler, cleaner and more effective technique which is suitable for large-scale industrial production. It can be carried out at room temperature without initiators and catalysts. In the case of in situ polymerization technique, the preparation of the gel electrolyte and the assembly of the batteries could be achieved in one step, improving the production efficiency and reducing the costs.

Depending on the method of packaging, batteries can have a coin, cylindrical, prismatic, or pouch shape. According to Li's work,²⁶ the large format pouch-type cells are widely used for high power applications. Compared with the coin-type half cells whose anode electrode is lithium metal, the pouch-type full cells are cheaper, safer, more practical and relevant to Li-ion battery devices.²⁷ Besides, the Al-plastic pouch has advantages of light weight, hermetic shielding, and freedom for use in various device designs.²⁸

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In this work, a novel GPE based on 3-(dimethylamino) propyl methacrylate (DMAPMA), poly(ethylene glycol) diacrylate (PEGDMA) and 1 M LiPF₆ liquid electrolyte solution was prepared by γ -radiation technique through a one-step synthesis method, called PDMP-Li GPE. The thermal stability and the mechanical properties of the resultant gels have been tested. And the ionic conductivity of the gel samples was studied by impedance spectroscopy. Finally, in order to confirm the possibility of PDMP-Li GPE usage in lithium-ion batteries, we tested its electrochemical properties in the pouch-type full cells.

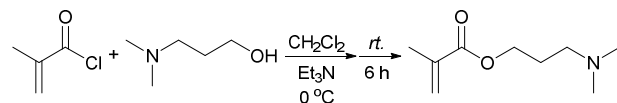
Experimental

Materials

2-Dimethylaminomethacrylate (98%), methacryloyl chloride (98%), lithium hexafluorophosphate (LiPF₆, 98%) and poly(ethylene glycol) diacrylate (PEGDMA, average Mn = 400) were bought from J&K and used without further purification. Liquid electrolyte (1 M LiPF₆ in EC/DEC 1:1 v:v) without any other additives (such as safety agents or fire retardant additives) was purchased from Tinci Company and used as supplied. Other chemicals were analytic-grade reagents obtained from Beijing Chemical Company and used as received.

Synthesis of 3-(dimethylamino) propyl methacrylate (DMAPMA)

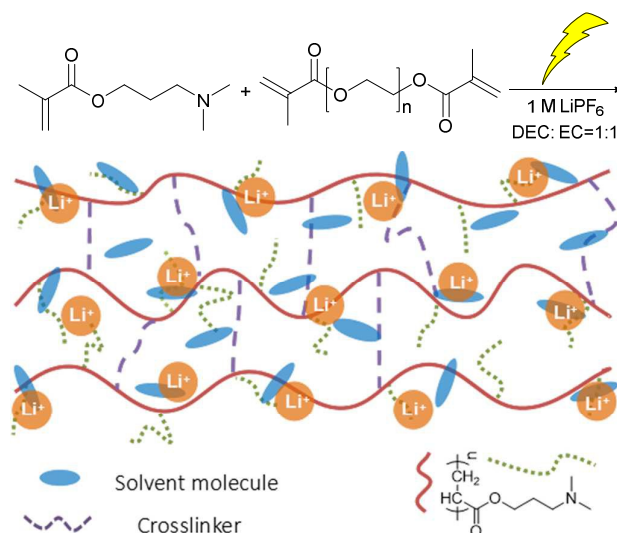
The 3-(dimethylamino) propyl methacrylate (DMAPMA) was prepared and purified according to the procedure published earlier by P. van de Wetering (Scheme 1).²⁹ Briefly, to an ice-cooled solution of DMAP-OH (3.60 mL, 30 mmol) in CH₂Cl₂ (300 mL) were added triethylamine (8.32 mL, 60 mmol) under magnetic stirring, then added methacryloyl chloride (5.38 mL, 54 mmol) by dropwise in 30 minutes. After stirred for 5 h at ambient temperature, the orange liquid was washed by saturated NaHCO₃ solution, then the organic layer was dried on Na₂SO₄ and filtered. The product was isolated by evaporation under reduced pressure, giving a 80% yield. ¹H NMR (400 MHz, CDCl₃, TMS standard, r.t.): δ (ppm) 6.10 (s, 1H, C=CH), 5.55 (s, 1H, C=CH), 4.20 (t, 2H, OCH₂), 2.37 (t, 2H, NCH₂), 2.24 (s, 6H, N(CH₃)₂), 1.95 (s, 3H, C=CCH₃), 1.85 (m, 2H, OCH₂CH₂CH₂N).



Scheme 1 The synthetic route of the 3-(dimethylamino) propyl methacrylate (DMAPMA).

Synthesis of PDMP-Li GPE

Synthesis route of PDMP-Li GPE is shown in Scheme 2. The concentrations of DMAPMA (monomer) and PEGDMA (crosslinker) were 0.75 mol L⁻¹ and 0.05 mol L⁻¹, respectively. The DMAPMA/PEGDMA were placed in a glass tube of 10 mm diameter and dissolved in the liquid electrolyte. The mixture was bubbled with nitrogen for 15 min. Finally, the tube was sealed and irradiated to form the gel with 10 kGy dose at a



Scheme 2 Schematic illustration of the preparation of PDMP-Li GPE.

dose rate of 80 Gy min⁻¹ at room temperature. The resulting gels were cut into cylindrical pieces and dried in the air at 25 °C for one day. For comparison, PDMAPMA gels (short for PDMP) were prepared under the same gelation condition only without the addition of LiPF₆.

Characterization of PDMP-Li GPE

Micro-FTIR and thermogravimetric analysis (TGA) were used to characterize the composition and thermal stability of the resultant gels. Micro-FTIR analysis was performed using a Nicolet (Magna-IR 750) spectrometer in the range of 5000-600 cm⁻¹. TGA was carried out using a TA Q600 thermal analyzer under a nitrogen atmosphere, over a temperature range of 25-600 °C with a heating of 5 °C/min. The gel fraction (GF) is defined as:

$$\text{Gel fraction (\%)} = \frac{W_g}{W_o} \times 100 \quad (1)$$

where W_g is the weight of the gel part and W_o is the original total dry weight.

The mechanical properties were measured using as-synthesized gel which was cut into cylindrical pieces ($d = 10$ mm), and then the compression strength test was performed by Instron 3365 material test system at room temperature.

The electrochemical measurements were carried out on an electrochemical instrument (CHI-660D, Shanghai). The ionic conductivity of the GPE was measured with a symmetrical cell SS/GPE/SS by EIS. The GPE was sandwiched between two SS discs (diameter $\varnothing = 16$ mm). The ionic conductivity of the GPE was calculated from the bulk electrolyte resistance (R) according to:

$$\sigma = \frac{l}{RS} \quad (2)$$

where l is the thickness of the GPE, and S is the contact area between GPE and SS disc. The bulk electrolyte resistance was obtained from the complex impedance diagram. In all the measurement for electrochemical impedance spectrum, the alternative current signal is from 100 kHz to 0.1 Hz with potential amplitude of 10 mV.

In situ synthesis and characterization of PDMP-Li GPE in pouch-type Li-ion cells

In situ synthesis and characterization of PDMP-Li GPE were carried out in LiFePO₄/graphite pouch-type cells. The production process of the pouch-type cells was similar to the commercial ones. The positive electrodes were obtained by coating the mixing slurry containing 92 wt.% commercial-grade LiFePO₄ active material, 3 wt.% Super P, 1 wt.% conductive graphite KS6 and 4 wt.% poly(vinylidene fluoride) (PVDF) onto an aluminium foil collector. The negative electrodes were prepared by coating the mixing slurry of 93 wt.% commercial-grade man-made graphite, 3 wt.% Super P and 4 wt.% poly(vinylidene fluoride) (PVDF) onto a copper foil. The separator was chosen commercial-grade. The negative electrode, positive electrode, and separators (Celgard 2325, USA, 25 μm in thickness) were subsequently rolled together to make a cell core. The cell core was dried at 85 °C overnight and put into cell shells made from aluminium film. The structure of pouch-type Li-ion cell and preparation process of PDMP-Li GPE was shown in Scheme S1. The injection of the excessive mixture liquid electrolyte (1 M LiPF₆ in EC/DEC 1:1 v:v) containing DMAPMA/PEGDMA (0.75 mol L⁻¹ and 0.05 mol L⁻¹, respectively) was carried out in an Ar-filled glovebox, after resting for 30 min, then the excess electrolyte and the formation gas were removed with a vacuum degassing machine to facilitate the diffusion of electrolyte into the cell core, finally remaining the content of electrolyte about 5 g/Ah. Subsequently, the cells were sealed with a sealing machine and the assembled cells were put in an oven (40 °C) for 24 h, ensuring that the electrolyte was fully absorbed.

Then the cells were irradiated with 10 kGy dose at a dose rate of 80 Gy/min at room temperature. In this process, the radiation-induced polymerization and crosslinking of DMAPMA and PEGDMA in the liquid electrolyte took place. Since the absorbed dose was only 10 kGy, during the irradiation, there was no significant change in the separators, which are mainly composed of polypropylene and polyethylene.^{30, 31} Meanwhile, the PDMP-Li GPE was prepared *in situ* inside the cells by one-step method.

The cell performances including charge-discharge curve and cyclic property were examined by using the charge-discharge testing device (NEWARE CT-3008-5V500 mA, Shenzhen, China). Cycle performance of the cells was evaluated at 0.1 C and 25 °C between 2.3 V to 3.6 V. All the cells were charged at room temperature by the constant-current/constant-voltage (CC-CV) protocol. That is, the cells were charged at a certain current rate until the voltage reached to a limited voltage, followed by holding the voltage until the current dropped to 10 mA.

Electrochemical impedance spectroscopy (EIS) was measured using a frequency response analyser (CHI-660D, Shanghai) with an electrochemical interface in the frequency range from 100 kHz to 0.1 Hz at an amplitude of 5 mV and 25 °C.

Morphology study of the electrode was observed using a scanning electron microscope (SEM, Hitachi S-4800, Japan), and the electrodes here were dried in vacuum at 60 °C for three days.

Results and discussion

Preparation and characterization of PDMP-Li GPE

It has been widely known that radiation-induced polymerization and crosslinking is an efficient and clean method, which needs no initiators and can be done at room temperature. In the process, the polymerization and crosslinking of the monomer is initiated by the radicals generated from the radiation. As shown in Scheme 2, a certain percentage of DMAPMA and PEGDMA were dissolved in the

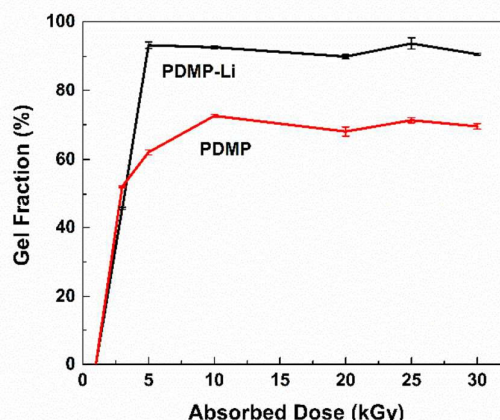


Fig. 1 The gel fraction of PDMP-Li GPE and PDMP gels as functions of absorbed dose.

liquid electrolyte, then after the system being irradiated, the DMAPMA monomer and the PEGDMA crosslinker formed an orange gel with the liquid electrolyte involved. The crosslinking network ensured the organic solvents won't leakage which improved the safety of the lithium-ion battery with the gel electrolyte.

Gel fractions (GF) of PDMP-Li GPE and PDMP gels as a function of absorbed dose are shown in Fig. 1. Similar to other gels, the formation of gels starts at a critical absorbed dose (gelation dose), and the gel fraction increases sharply with the increasing absorbed dose in a narrow range and then levels off.³² It was found that the PDMP-Li GPEs were synthesized at lower absorbed dose and the gel fractions were much higher compared with that of PDMP gel without LiPF₆. It is worth noting that high GF could be gained by relatively low absorbed dose, which brings advantages for avoiding the radiation side effects. And the radiation-induced polymerization

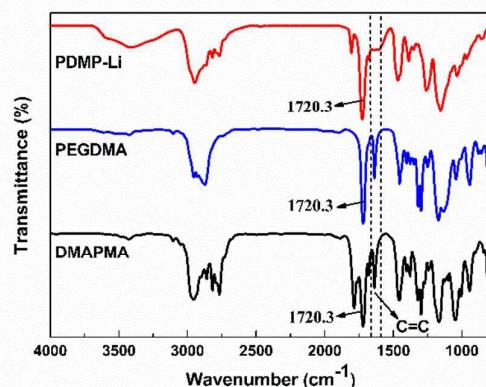
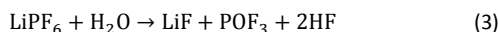


Fig. 2 FTIR spectra of PDMP-Li GPE, PEGDMA crosslinker, DMAPMA monomer.

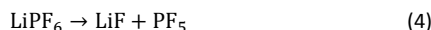
and crosslinking was much more efficient for PDMP-Li GPE system, obtaining GF of ca. 93% at only 5 kGy, maybe the LiPF₆ inside the system as an additive improved the formation of crosslinking network.³³

FTIR and TGA were carried out to characterize the chemical structure and thermal stability of the resultant gels, respectively. Herein, the FTIR spectra of PDMP-Li GPE, PEGDMA crosslinker and DMAPMA monomer are shown in Fig. 2. For PEGDMA crosslinker and DMAPMA monomer, the peaks of 1637.3 cm⁻¹ and 1720.3 cm⁻¹ are ascribed to the functional groups of C=C and C=O, respectively. Compared with the FTIR spectra of PEGDMA and DMAPMA, it can be found that PDMP-Li GPE remained the absorption peak at 1720.3 cm⁻¹ for C=O, while the absorption peak of C=C group disappeared. The FTIR results suggest that the crosslinking network is obtained through opening the C=C double bonds in the monomer and crosslinker, while the main functional groups are remained.

Fig. 3 shows the TGA curves of LiPF₆, liquid electrolyte (1 M LiPF₆ in EC:DEC 1:1 v:v), PDMAPMA gel and PDMP-Li GPE. By comparing the weight loss as a function of temperature, the order of thermal stability is liquid electrolyte < LiPF₆ < PDMP-Li GPE < PDMAPMA gel. It is well known that the commercial LiPF₆ is very unstable at elevated temperature as shown in curve 3. LiPF₆ decomposes in two steps, the first decomposition step is attributed to the traces of water and its decomposition occurs according to reaction 3.³⁴



The main decomposition starts at 85 °C, and its decomposition path is a simple dissociation producing lithium fluoride (LiF) as solid and PF₅ as gaseous products according to reaction 4.³⁴



In curve 4, the liquid electrolyte shows the worst thermal stability: the organic solvents start to volatilize at the beginning,

and when the temperature arrives at 80 °C, the liquid electrolyte remains 80% of its initial weight. A decomposition at temperature above 80 °C may occur according to reaction 4. As shown in curve 1, pure PDMAPMA gel is thermally stable up to 250 °C, the first decomposition is related to the dimethylamino in the structure of PDMAPMA. Another decomposition is observed in the range of temperature from 350 °C to 400 °C, probably due to the carbonization loss of the backbone segments.³⁵ Compared with curve 3 and curve 4, it is obvious that PDMP-Li GPE expresses more excellent thermal stability (curve 2), i.e. before 100 °C, PDMP-Li GPE does not lose any weight, indicating the LiPF₆ and organic solvents in the gelled network are more stable; for the temperature range of complete thermal decomposition, all the systems are similar. Although the PDMP-Li GPE starts to decompose at 120 °C, which is at the same level with others' works,^{17, 19} and this degradation temperature is still high enough for the practical use.

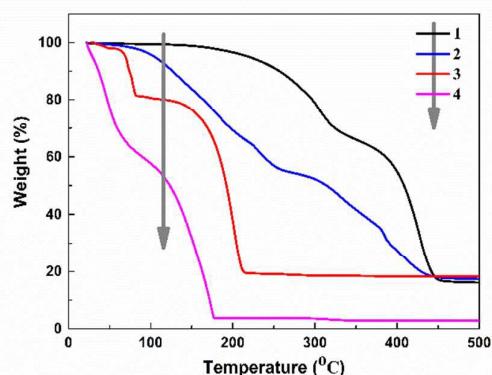


Fig. 3 The TGA graph of the samples; curve 1: PDMAPMA gel; curve 2: PDMP-Li GPE; curve 3: LiPF₆; curve 4: liquid electrolyte.

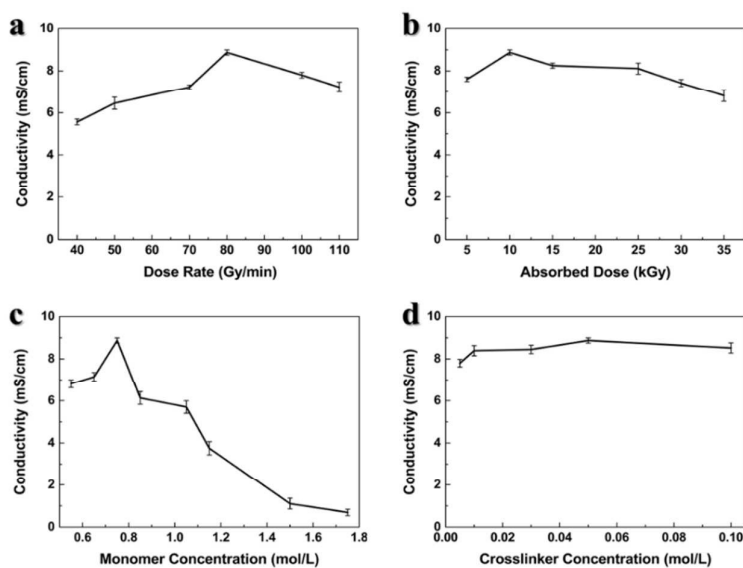


Fig. 4 The ionic conductivity of PDMP-Li GPE as functions of a) dose rate (the absorbed dose was 10 kGy, the concentration of DMAPMA was 0.75 mol/L, the concentration of PEGDMA was 0.05 mol/L); b) absorbed dose (the dose rate was 80 Gy/min, the concentration of DMAPMA was 0.75 mol/L, the concentration of PEGDMA was 0.05 mol/L); c) DMAPMA concentration (the dose rate was 80 Gy/min, the absorbed dose was 10 kGy, the concentration of PEGDMA was 0.05 mol/L); d) PEGDMA concentration (the dose rate was 80 Gy/min, the absorbed dose was 10 kGy, the concentration of DMAPMA was 0.75 mol/L).

The ionic conductivity of the GPEs plays an important role on the electrochemical performance. In Fig. 4, we studied the influence factors against the ionic conductivity of PDMP-Li GPE, and determined the optimal synthesis conditions. When the absorbed dose and dose rate are 10 kGy, 80 Gy/min, the concentration of DMAPMA and PEGDMA are 0.75 mol L⁻¹, 0.05 mol L⁻¹, respectively, the highest ionic conductivity of the resultant PDMP-Li GPE reaches 8.88×10⁻³ S cm⁻¹ at room temperature, which is higher than the most results that have been reported in literatures and competitive to the liquid electrolyte that has been widely used in lithium-ion battery. As the ion transport in the LiPF₆ EC/DEC solution is mainly achieved by Li⁺ ions, the high conductivity in PDMP-Li GPE is caused

by the higher lithium-ion concentration per repeating unit. Huang³⁶ reported that the Li⁺ ions within the GPEs were mainly solvated by the C=O because of their high electronegativity which dissociated lithium salt and hindered the movement of large PF₆⁻ anions. Therefore, because of the strong interaction, the carbonyl oxygen can separate the Li⁺ and PF₆⁻ ions in the PDMP-Li GPE which may prevent the reunion between Li⁺ and PF₆⁻ ions concurrently and create more free Li⁺ ions. Fig. 4a and Fig. 4b indicated that when the samples were irradiated for a long time, the high ionic conductivity may be sacrificed, but all the order of magnitude of the ionic conductivity was about 10⁻³ S cm⁻¹ which is still higher than most reports as shown in Table 1.

Table 1 Ionic conductivities of various GPEs reported in literatures.

| Gel composition (polymer/electrolyte) ^a | Ionic conductivity [mS/cm] | Ref. |
|---|-------------------------------|-----------|
| PVDF-LiPAAOB/LiPF ₆ -EC-DMC-EMC | 0.35 (25 °C) | 11 |
| OEGMA-CCMA/LiPF ₆ -EC-DMC | 2.30 (25 °C) | 17 |
| BC-PVDF/LiTFSI-EC-DEC | 4.20 (30 °C) | 18 |
| P(AN-VAc)-PMMA/LiPF ₆ -EC-DMC-DEC | 3.50 (30 °C) | 22 |
| CA-PLLA-HNT/LiPF ₆ -EC-DMC | 1.52 (25 °C) | 25 |
| PVDF-PPC/LiPF ₆ -EC-DMC | 4.05 (30 °C) | 36 |
| P(MMA-AN-VAc)/LiPF ₆ -EC-DMC-DEC | 3.48 (25 °C) | 37 |
| PDMPMA/LiPF ₆ -EC-DEC | 8.88 (25 °C) | this work |

^aLiPF₆, lithium hexafluorophosphate; LiTFSI, lithium bis(trifluoromethane sulfonimide); EC, ethylene carbonate; EMC, ethyl methyl carbonate; DEC, diethyl carbonate; DMC, dimethyl carbonate; PVDF, poly(vinylidene fluoride); LiPAAOB, lithium polyacrylic acid oxalate borate; OEGMA, oligo(ethylene glycol) methyl ether methacrylate; CCMA, cyclic carbonate methacrylate; BC, boron-containing crosslinker; P(AN-VAc), poly(acrylonitrile-co-vinyl acetate); CA, cellulose acetate; PLLA, poly-L-lactic acid; HNT, halloysite nanotube; PPC, poly(propylene carbonate); P(MMA-AN-VAc), poly(methyl methacrylate-co-acrylonitrile-co-vinyl acetate);

Fig. 4c shows the ionic conductivity of PDMP-Li GPE as a function of DMAPMA concentration. The ionic conductivity first increased with increasing concentration of DMAPMA due to the denser gel network and high degree of gelation which remained the liquid electrolyte inside the gel network, and the maximum ionic conductivity was observed at the DMAPMA concentration of 0.75 mol L⁻¹. Then, the ionic conductivity began to decrease with further increase of the DMAPMA concentration, because the much denser network caused less liquid electrolyte inside the gel, then led to the decrease.³⁸

According to Fig. 4, the monomer concentration influences the ionic conductivity most. So in Fig. 5, we observed the stress-strain curves of the resultant PDMP-Li GPE against the monomer concentration at room temperature. Usually, for solid state materials, after soaked liquid, the mechanical property becomes worse.³⁹ The compressive stress and the elastic modulus increased with DMAPMA concentration, while compressive strain decreased. The order of magnitude of compressive stress was about 1 MPa, among the monomer concentration from 0.6 mol L⁻¹ to 3.0 mol L⁻¹. The reasonable explanation for this phenomenon is that the higher the monomer concentration, the denser the gel network, which means less liquid electrolyte was absorbed in the network. This makes the gel tougher and hard to be compressed. As we can see,

when the ionic conductivity reaches 8.88×10⁻³ S cm⁻¹, the compressive stress is about 0.4 MPa which is high enough to meet

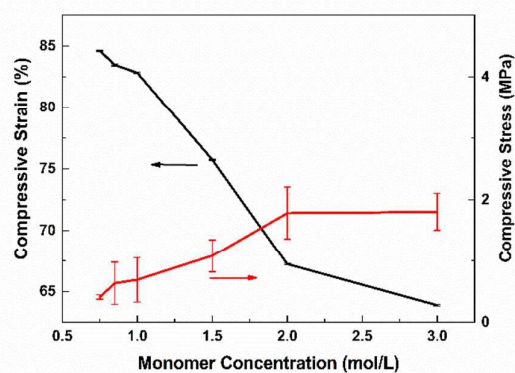


Fig. 5 The compressive strain and compression stress of PDMP-Li GPE as functions of DMAPMA concentration (synthesis condition: dose rate: 80 Gy min⁻¹; absorbed dose: 10 kGy; crosslinker concentration: 0.05 mol L⁻¹).

the needs of flexible batteries.

In situ synthesis and properties of PDMP-Li GPE in pouch-type Li-ion cell

ARTICLE

Journal Name

From the above discussion, it can be notified that the PDMP-Li GPE exhibited good thermal stability, excellent mechanical strength and especially presented high ionic conductivity around $8.88 \times 10^{-3} \text{ S cm}^{-1}$. To exam the actual application effort of the PDMP-Li GPE on the lithium-ion batteries, the LiFePO_4 electrode was chosen as cathode and the $\text{LiFePO}_4/\text{PDMP-Li GPE/graphite}$ pouch-type cells were assembled. For comparison, the liquid-state $\text{LiFePO}_4/\text{graphite}$ cells were also assembled by using Celgard 2325 porous membrane as separator.

In order to prove that PDMP-Li GPE was truly generated inside the pouch-type full cells, we removed the cell's shell and observed that there was few orange gel on the aluminium film. The thickness of PDMP-Li GPE was ca. 25 μm . We compared the SEM of the electrodes before and after radiation in Fig. 6. It can be seen from Fig. 6a and Fig. 6c that the surface of untreated electrodes was rough and the morphology of the electrodes could be seen clearly. However, the surface of the irradiated electrodes in Fig. 6b and Fig. 6d became extremely flat and seemed like the surface was covered evenly by the gelled layer which confirmed the PDMP-Li GPE truly existed.

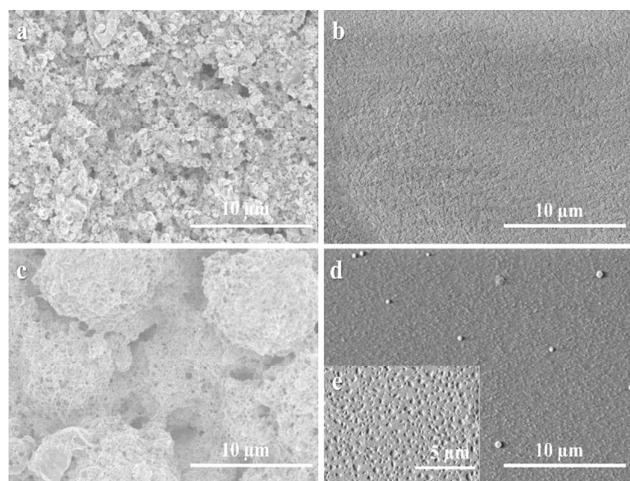


Fig. 6 The SEM images of the electrodes inside the pouch-type full cell with PDMP-Li GPE. a) untreated anode; b) anode after radiation; c) untreated cathode; d) cathode after radiation; e) the obtained cathode d) was washed with DEC several times, and dried under vacuum.

It was found that there were some white spots on the surface of electrode in Fig. 6d. We speculate that LiPF_6 may be precipitated on the surface of GPE. In order to confirm our thought, the obtained cathode in Fig. 6d was further washed with DEC several times and died under vacuum. As shown in Fig. 6e, it can be seen that there were some small holes in the surface uniformly which confirmed that the holes were produced in situ after the dissolution of LiPF_6 . Therefore, the liquid electrolyte with DMAPMA and PEGDMA additives was totally gelled uniformly to form PDMP-Li GPE after radiation in the pouch-type full cells and the high ionic conductivity may be attributed to the evenly distributed LiPF_6 within the GPE.

Fig. 7a presents the cyclic performances of the $\text{LiFePO}_4/\text{graphite}$ pouch-type cells. The cells were cycled with a constant current of 0.1 C rate between 3.6 and 2.3 V. After 50 cycles, the capacity retention of the pouch-type cell with PDMP-Li GPE achieved a value of 86%, slightly lower than that of the pouch-type cell with liquid electrolyte, which retained 93% of its initial discharge capacity.

These results are in line with the EIS spectra as shown in Figure S1. The decrease in discharge capacities of the pouch-type cell with

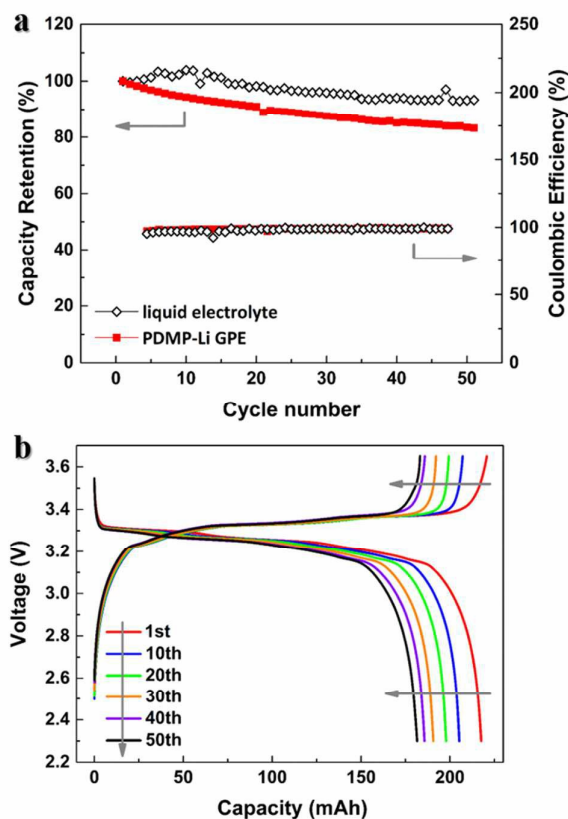


Fig. 7 a) Cycling performances of the assembled $\text{LiFePO}_4/\text{graphite}$ pouch-type cells with PDMP-Li GPE and liquid electrolyte (1 M LiPF_6 in EC:DEC 1:1 v:v) under 0.1 C rate in the voltage range of 2.3 V to 3.6 V at room temperature. b) detailed charge/discharge curves of the pouch-type cell with PDMP-Li GPE.

PDMP-Li GPE may occur due to the formation of passive layers over the electrode surface during cycling. Taking the pouch-type cell with PDMP-Li GPE for example, the detailed charge and discharge curves are demonstrated in Fig. 7b. The characteristic voltage plateau of LiFePO_4 at about 3.3 V was quiet stable. Therefore, the application of PDMP-Li GPE in pouch-type full cells will not sacrifice the cycling stability of LiFePO_4 cathode, resulted from its high ionic conductivity.

Conclusions

PDMP-Li GPE consisted of 1 M LiPF_6 EC/DEC (1:1 in volume) and DMAPMA gel was synthesized successfully by one-step method with γ -radiation induced polymerization and crosslinking. The resultant PDMP-Li GPE exhibited high ionic conductivity around $8.88 \times 10^{-3} \text{ S cm}^{-1}$, good thermal stability, excellent mechanical integrity and strength to allow the safe operation in lithium-ion batteries. After 50 cycles, the assembled $\text{LiFePO}_4/\text{PDMP-Li GPE/graphite}$ pouch-type cell showed a very stable discharge behavior and little capacity loss under current constant voltage conditions, at rate of 0.1 C and 25 $^\circ\text{C}$. Considering these results, the cell with PDMP-Li GPE appears as a promising candidate for lithium-ion batteries.

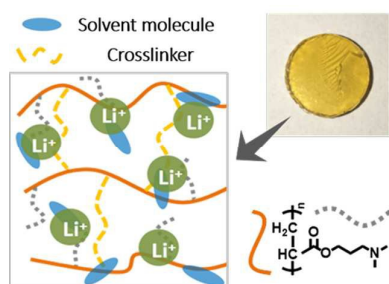
Acknowledgements

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Table of Contents Entry



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