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Elastomers Uploaded Electrospun Nanofibrous Membrane as Solid State Polymer Electrolytes for Lithium-ion Batteries

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Abstract: A new class of thin, safe and foldable solid composite electrolytes are produced by uploading 3-arm poly-(methoxy-poly (ethylene glycol) methacrylate) (3PEG) on three different electrospun membranes, poly(vinylidenefluoride-co-hexafluoropropylene) (P(VdF-HFP)) membrane, 3PEG co-sprayed P(VdF-HFP) membrane (3PEG/PHP) and polyethylene terephthalate membrane (PET), respectively, as named 3PEG@PHP, 3PEG@3PEG/PHP and 3PEG@PET. The electrospun membranes serve as skeleton to enhance mechanical strength of composite electrolytes, while 3PEG filled in micropores of the matrix affords ion transport carrier. Among these composite electrolytes, the one based on PET electrospun membrane exhibits a high ionic conductivity of 5.9×10^{-4} S cm⁻¹ at 100 °C because of its least shrinkage rate with high filling loading of 3PEG. The decomposition potential of the composite electrolytes is above 4.5 V at 100 °C. Successful charge and discharge cycling of 3PEG@PET based all solid state lithium ion battery have maintained with the initial discharge capacity of 137.6 mAh g-1 at 0.1C, which proves that they are deal candidates for all solid state rechargeable lithium ion battery.

Keywords: Composite electrolytes; Electrospun membranes; Elastomers; High temperature; Lithium-ion battery

1. Introduction

Rechargeable lithium-ion batteries (LIBs), being used in emerging electric vehicles, all kinds of portable electronics and so on, have drawn world wide attention due to its high energy density, excellent cycle life and low self-discharge rate.¹⁻³ Strong market demands the need for development of projected and advanced electrolytes for lithium-ion batteries.⁴ The commercial electrolytes of LIBs are usually liquid solutions mainly composed of carbonate solvents and lithium salt, such as propylene carbonate (PC), diethyl carbonate (DEC), dimethyl carbonate (DMC) and ethylene carbonate (EC). Unfortunately, these solvents exhibit high vapor pressures at elevated temperatures (>60 °C), leading to a tendency of leakage and flammable. At the same time, batteries able to work at relatively high temperature are needed for some specific applications, such as petroleum exploration.⁵⁻⁸ Therefore, developing safe and efficient polymer electrolytes without any liquids, i.e. solid polymer electrolytes (SPEs), is urgently in demand.

To date, poly (ethylene oxide) (PEO) based electrolytes are widely studied because of their effective ion solvating properties, but their conductivities (about 10^{-7} S cm⁻¹ at ambient temperature) are far below the level of ionic conductivity for practical applications $(10^{-3} S \text{ cm}^{-1})$. Branched polymer electrolytes have been considered as one of the most promising candidates because they are intrinsically resistant to crystallization, which is advantageous in improving their conductivities. For example, branched PEO with polyimide main chain and methoxy-poly (ethylene glycol) methacrylate side chains achieves a higher ionic conductivity $(10^{-5} - 10^{-6} \text{ S cm}^{-1})$ than linear PEO-based polymer at ambient temperature.^{9,10} Previous studies have reported the synthesis of 3-arm branched poly (poly-(ethylene glycol) methyl ether methacrylate) (3PEG) via atom transfer radical polymerization. It was noted that 3PEG has an amorphous state at room temperature due to the branched structure. This unique structural feature of 3PEG can provide enhanced ionic conductivity when employed as a polymer matrix for $SPEs$.¹¹ But the application of branched PEO-based polymers for commercial use is staggered due to their poor mechanical properties,

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especially at high temperature. When the temperature goes beyond 100°C, the fluidity of them lead to internal short-circuit failures between electrodes because of the low melting temperature of PEO-based polymer electrolyte.

One favorable solution to this problem involves the use of electrospun matrix to isolate electronic flow between the cathode and anode to prevent physical contact in the inner of batteries by decreasing fluidity of the polymer electrolyte at high temperature as well as offsetting their poor mechanical properties. Electrospun membranes possess remarkable characteristics such as large surface-to-volume ratio, abundant pore number as well as chemical compatibility with Li electrode, among which P(VdF-HFP) and PET are especially preferred because of their high dielectric constant, good thermal, electrochemical stability and affinity to electrolytes.¹²⁻¹⁶

In this study, thin, deformable, safe and high-temperature-tolerant novel composite electrolytes are obtained by combining 3-arm branched poly-(methoxy-poly (ethylene glycol) methacrylate) (3PEG) with three electrospun matrices including P(VdF-HFP) membrane, 3PEG cladding P(VdF-HFP) membrane (3PEG/PHP) and poly (ethylene terephthlate) membrane (PET), respectively. The porous substrates act as a mechanical framework, while 3PEG is the ion transport carrier. The composite electrolytes exhibit good thermal and mechanical properties, without impairing advantageous features such as high ionic conductivity of 3PEG. Electrochemical test results reveal that the operating temperature for the present composite electrolyte is up to 100°C.

2. Experimental section

2.1. Materials

 $3PEG$ was synthesized by the method shown in our previous work.¹¹ Poly (vinylidene) fluoride-co-hexafluoropropylene) (P(VdF-HFP)), Mn= 4×10^5 g mol⁻¹, Aldrich) and poly (ethylene terephthalate) (PET, inherent viscosity=0.8 dl g⁻¹, Yuanfang Co., Ltd. Shanghai) were dried in a vacuum oven at 60° C for 24 h before used. Lithium perchlorate $(LiClO₄)$ was dried in a vacuum oven and transferred to argon-filled glove box with oxygen and moisture level ≤ 0.1 ppm. LiFePO₄, PVdF binder, carbon black and lithium metal anode were chemically pure and purchased from Damao Chemicals, Tianjin, China. Trifluoroacetic acid, acetone, N, N-dimethylformamide, dichloromethane, acetonitrile were obtained from China Medicine Co., Ltd., and used as received.

2.2 Preparation of nanofibrous matrices

P(VdF-HFP) nanofibrous membranes were prepared by electrospinning. A 16wt. % solution of P(VdF-HFP) for electrospinning was prepared by dissolving P(VdF-HFP) in a mixture of acetone and N, N-dimethylformamide (DMF) (3/1, *V*/*V*) by mechanical stirring over night. The solution was taken in the syringe and delivered with a flow rate of 1.2 ml h^{-1} . Electrospinning progress was conducted at an electric voltage of 20 kV at room temperature with the above solution, the distance between target drum and the syringe needle tip (inner diameter 22μ m) was 15 cm.

PET nanofibrous membranes were obtained similarly to P(VdF-HFP) nanofibrous membranes. 10wt. % solution of PET dissolving in a mixture of trifluoroacetic acid (TFA) and dichloromethane $(3/2, V/V)$ was electrospun at the applied voltage of 25 V at room temperature with a flow rate of $3 \text{ ml } h^{-1}$. The grounded collector was located at a fixed distance of 15 cm from the needle tip.¹⁷

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To ensure the surface of each P(VdF-HFP) nanofiber being coated with 3PEG, the 3PEG/PHP nanofibrous membranes were prepared by combining electrospinning with electrospraying. Two-needle electrospinning/electrospraying apparatus was set up. P(VdF-HFP) solution was fed in one syringe pump with the same parameters used for electrospinning while 3PEG solution (6wt. %, acetonitrile) in another syringe parallel to the former one was provided for spraying. Two needles were arrayed closely and parallel in the direction vertical to the axes of rotating cylinder with the one for spray slightly higher than the other one, thus 3PEG coated P(VdF-HFP) nanofibers were formed, simultaneously. This method is analogous to coaxial electrospinning.^{18, 19} Finally, three fabricated nanofibrous matrices dried under vacuum at 60°C for 24h and used for characterization and further studies.

2.3 Preparation of composite electrolytes

A 3PEG and LiClO₄ solution ($[O]/Li^+$] ratio=20) with predetermined amounts of 3PEG and LiClO4 dissolved in anhydrous acetonitrile was stirred for several hours to form homogeneous solution. After immersing nanofibrous membranes (3PEG@PHP, 3PEG@3PEG/PHP and 3PEG@PET) into the mixture for 6 hours, the solvent-containing composite electrolyte films yielded. To obtain the designed composite electrolyte membranes, the as prepared films were finally dried under vacuum at 45 °C for 24 h. The manufacturing processes are showed in **Scheme 1**.

2.4 Coin cell assembly

LiFePO $_4$ /Li half cells were fabricated with 2025-coin type cells in the argon-filled glove box using the 3PEG@PET as electrolyte. LiFePO₄ electrodes were prepared by mixing the LiFePO₄ powder, carbon black and PVdF in the ratio of $80:10:10$ by wt. in N-methylpyrrolidone (NMP) solvent. Mixed slurry was then coated onto aluminum foil and dried at 120 \degree C for 6 h. The assembly of cells were carried out in argon-filled glove box with a moisture level <10 ppm.

2.5 Characterizations

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Morphology of nanofibrous matrices and composite polymer electrolyte membranes were observed by scanning electron microscopy (SEM, FEI Quanta 200SEM) operating at 20 kV. Thermal gravimetric analysis (TGA, Perkin-Elmer TGA 7) with the heating rate of 15 $^{\circ}$ C min⁻¹ under nitrogen atmosphere is used to measure the thermal degradation temperatures (T_d) . Glass transition temperatures $(T_g s)$ were measured with differential scanning calorimeter (DSC, TA instrument Q2000) at heating rate of 10 °C min⁻¹ under nitrogen atmosphere. The *T*_gs of samples were obtained using mid-point method on the second heating curve. The mechanical measurement of the composite electrolyte membranes was carried out on CMT-8000 mechanical testing apparatus (SANS Corp., Shenzhen, China) with specimen width of 1.0 cm. The ionic conductivities of all the prepared composite polymer electrolyte were gauged by electrochemical impedance spectroscopy (EIS) using Zahner Zennium electrochemical analyzer (Germany) with SPE sample membranes sandwiched between two stainless-steel (SS) electrodes and measured at amplitude of 10 mV under the frequency of 1 Hz to 100 kHz. The samples were thermally equilibrated at each temperature for 30 minutes to ensure the precision of every test dot. Ionic conductivity of SPE can be calculated from following Eq. (1),

$$
\sigma = \frac{L}{R_b S} \tag{1}
$$

where R_b represents the bulk resistance, L is the thickness and S stands for the area of the sample. The electrochemical stability of the samples was investigated by linear sweep voltammetry (LSV). All experiments were carried out at a sweep rate of 5 mV $s⁻¹$ in the voltage range of 2.0 -7 V (*vs.* Li⁺/Li) at 100 °C. Cycling performance tests of the $Li/3PEG@PET/LiFePO₄$ cells were conducted in an automatic galvanostatic charge–discharge unit, Neware BTS (Shenzhen, China), over the range of 2.5-4 V at 80 °C and 100 °C.

3. Results and Discussion

3.1 Morphological analysis and thermal behaviors

The 3PEG filled electrospun membranes were carried out in anhydrous acetonitrile solution at room temperature. Digital pictures of 3PEG elastomer and composite electrolytes are shown in **Figure 1**. Compared with 3PEG elastomer, the macro morphology of 3PEG@PET with 3PEG directly filled in the PET electrospun substrate shows that the composite electrolyte membranes are deformable and remain constant, which can satisfy the application in the foldable lithium-ion battery. The images of P(VdF-HFP), 3PEG/PHP and PET nanofibrous membranes prepared by electrospinning are presented in **Figure 2**. It can be seen that the P(VdF-HFP) and PET nanofibers randomly dispersed in the membranes with average fiber diameter and smooth surface. Though the 3PEG/PHP nanofibers have relatively rough morphology and large diameter, the interconnected open pore structure of the 3PEG-clad electrospun membranes still remained unmodified (**Figure 2**). Fig. 2D-F shows the morphology of 3PEG@PHP, 3PEG@3PEG/PHP and 3PEG@PET. After immersing in the 3PEG and $LiClO₄$ acetonitrile solution, 3PEG was filled in the micropore of the electrospun membranes. Among the three composite electrolytes, 3PEG@PHP shows least filling rate due to their poor compatibility with 3PEG. In virtue of the 3PEG coated on the surface of the nanofibers, $3PEG@3PEG/PHP$ membrane possessed better uniformly surface without any micropore. So did 3PEG@PET, the micropores were filled with 3PEG continuously. **Figure 3** presents the macro-morphology and shrinkage of three composite electrolytes based on different electrospun membranes by measuring the (area-based) dimensional change. The composite electrolytes based on P(VdF-HFP), 3PEG@PHP and 3PEG@3PEG/PHP show large shrinkage and rough surface, while the dimensional change of the 3PEG@ PET appears to be negligible. The results imply that the 3PEG@PET electrolyte exhibits good shape retention and flat surface. Thermal stability of the electrolytes was first evaluated by TGA (**Figure 4A**). The weight loss occurred after 250 \degree C for all of the composite electrolytes (3PEG@PHP,

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3PEG@3PEG/PHP and 3PEG@PET), suggesting that the prepared electrolytes are stable up to 250 \degree C, which is high enough for use in lithium ion batteries. The thermal properties of composite electrolytes doped with lithium salt are also determined by DSC (**Figure 4B**) and samples are first heated to 150 °C to remove thermal history. The glass transition temperatures (T_gs) of all the electrolytes are far below room temperature, which means that the 3PEG chain shows good segment movement ability regardless the filler effect in electrospun membrane. 3PEG@PHP electrolyte shows the highest T_g value, possibly due to the poor compatibility between P(VdF-HFP) and 3PEG which inhibits the movement of the 3PEG segment.

3.2 Mechanical and electrochemical properties

Impedance spectroscopy is employed to acquire the ionic conductivity of electrolytes within the temperature ranging from 30° C to 100° C by using an AC impedance analyzer with the composite polymer electrolytes sandwiched between a stainless steel blocking electrode. The ionic conductivities of electrolytes based on different thickness of the electrospun matrix are presented in **Figure 5.** It can be seen that the ionic conductivity values reduce with the increase of thickness of the electrospun substrates, which is ascribed to the ionically nonconductive of electrospun nanofiber skeleton. Composite electrolyte membranes can hardly form independent films when the thickness of substrates decline to 10 μ m. A trade-off between ionic conductivity and mechanical properties must be made, so 20 µm is a good choice to achieve win-win composite electrolytes. Inset in **Fig. 5** gives the typical stress–strain curves for 3PEG@3PEG/PHP and 3PEG@PET respectively. It can be seen that the tensile strength of 3PEG@3PEG/PHP is 6.0 MPa, with an elongation at break of 102% and the tensile strength of 3PEG@PET is 6.6 MPa, with an elongation at break of 83%. The result suggests that the as prepared composite electrolytes have considerable tensile strength and good property of elongation. The temperature dependent ionic conductivity of the composite electrolytes are also measured and compared with that of pure 3PEG electrolyte. **Figure 6A** shows that the conductivities rise with the elevated temperature over a wide range of temperatures. The conductivity of the

composite electrolytes follows the order: $3PEG@PET > 3PEG@3PEG/PHP > 3PEG@3$ 3PEG@PHP both at ambient temperature and elevated temperature, which is the reverse order of T_g (**Table 1**). Since the ion conductivity occurs mainly through the amorphous region in the 3PEG matrix, the increase of the ionic conductivity for the composite polymer electrolytes mainly relates to the movement of the segment.²⁰3PEG@PET exhibits the highest conductivity value, this coincides with its significantly lowest T_g (-32.06 °C). The low ionic conductivity of 3PEG@PHP electrolyte film should also be ascribed to the poor compatibility of the 3PEG with the P(VdF-HFP) matrix, resulting in few 3PEG in the micropore of the nanofibrous membranes, which corresponds directly with that estimated by SEM image.^{21,22}

For a solid polymer electrolyte system, the temperature dependent ionic conductivity obeys the Vogel–Tamman–Fulcher (VTF) equation

$$
\sigma(T) = AT^{-\frac{1}{2}} \exp\left(-\frac{E_a}{R(T - T_0)}\right) \tag{2}
$$

Where σ is the ionic conductivity at different temperature, Λ is a frequency factor which often relates to the number and mobility of charge carriers, and E_a is the activation energy, R is the perfect gas constant, $T₀$ is the Vogel temperature at which relaxation times become infinite or the mobility of ions goes to zero. According to the literature, T_0 is 50 K below the glass transition temperature on polyether-based polymer electrolytes.²³ The VTF equation predicts that a plot of $\ln(\sigma T^{1/2})$ vs. $1/(T-T_0)$ should be linear, which is exactly observed in **Figure 6B**. The fitting results are listed in **Table 1**. *Ea* values of 3PEG and composite electrolytes vary from 0.056 eV to 0.072 eV, respectively, which are in agreement with the values for the amorphous state composite polymer electrolyte reported.²⁴⁻²⁶ When considering polymer electrolytes acting as electrolyte as well as separator, ionic conductance normalized by sample thickness is more important in determining cell performance than ionic conductivity.²⁷ Due to short ion diffusion channel, thin polymer electrolytes are advantageous in delivering high ionic conductance. Previous results (**Figure 5**) show

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that the electrospun matrix with the thickness about 20 μ m is the best choice taking both ionic conductivity and the morphology of the composite electrolytes into account. The thickness of the composite electrolytes, approximately 30μ m, is lower than 3PEG electrolyte $(\approx 100 \mu \text{ m})$ (Figure 6C) highlighting the advantages of thin composite electrolytes on the ionic conductance. Contrary to the results of ionic conductivity, 3PEG@PET exhibits higher ionic conductance than pure 3PEG electrolyte. Ionic conductance was calculated by Eg. (3):

$$
G = (\sigma A)/l \tag{3}
$$

The higher ionic conductance of electrolytes has been proved to be beneficial for circulation property of cell. 28

The characteristic EIS of 3PEG@PET at different temperatures from 30 °C to 100 °C are presented in **Figure 7**. The Nyquist plot is a slanted line without semicircle in the high-frequency. This owes much to the low T_g of composite electrolytes which ensure segment's excellent mobility and short relaxation time of the high-frequency response.²⁹ The semicircle replaced by the inductive response below the real axis is another reasonable possibility $.^{30}$

It is important to investigate the electrochemical stability of the electrolytes for practical battery applications within the operation voltage of the battery system. The electrochemical stability windows of 3PEG@3PEG/PHP and 3PEG@PET electrolytes are studied using linear sweep voltammetry with lithium foil as both the Counter and reference electrodes and stainless anodic region before observing appreciable current. The decomposition voltage of the polymer electrolyte was determined by the steel as the working electrode at 100 °C. The results are shown in Figure 8, in which the electrolyte was swept from 2 V to 7 V (versus Li/Li^{+}) at a constant rate of 5 mV s^{-1} . We notice that the electrochemical decomposition potentials of 3PEG@3PEG/PHP and 3PEG@PET are about 4.8 V and 4.9 V in the intersection of the line fitting the steep current increase and the voltage axis, respectively.³¹

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3PEG' s tightly trapping in the pores of the electrospun membrane contributes to the good oxidative stability of the composite electrolytes. Thus, this interaction will contribute to the high electrochemical stability window.³² That is similar to the value reported by Zhang et al.³³ for the system PEO_{16} -LiClO₄-0 wt. % MLA. Also Kang et al.³⁴ found that the electrochemical stability seemed almost similar between the linear and multi-armed electrolyte. Good electrochemical stability of the electrolyte at elevated temperature make the electrolyte as a potential candidate for lithium-ion batteries operated at a high voltage.

In order to evaluate the electrochemical performance of 3PEG@PET as electrolyte in lithium-cells, coin-type cells were fabricated with Li as anode and $LiFePO₄$ as cathode. The assembled cells were subjected to cycle tests. A weak Faraday current can be observed between 4 V and the decomposition potential for the electrolyte. For safety considerations, 4 V was set as the cut-off voltage for $Li/3PEG@PET/LiFePO₄$ battery cycling tests. The second charge-discharge profiles of Li/3PEG@PET/LiFePO4 cells at 80 °C and 100 °C are showed in **figure 9A**. Cells were cycled under a voltage range of 2.5-4 V at 0.1C. Higher temperature (100 °C) discharge capacity of the cell is 137.6mAh g^{-1} , which is significantly higher than that at 80 °C (123.5 mAh g^{-1}). This owes to the advanced segment movement of solid-state electrolyte and increased Li-ion mobility in LiFePO₄/FePO₄ mixed phase at elevated temperature.³⁵ The constant potential region around 3.4 V versus Li/Li^{+} at 100 °C was observed for the charge–discharge curves, suggesting that reversible charge/discharge reactions proceeded. **Figure 9B** presents discharge capacity of Li/3PEG@PET/LiFePO₄ upon cycling at 100 °C with a capacity retention exceeding 95.7 mAh g^{-1} for 50 cycles. The formation of a solid electrolyte interphase (SEI) and oxidability of lithium perchlorate may ascribe to the capacity decrease, leading to the discharge capacity of Li/3PEG@PET/LiFePO₄ cell lower than the theoretical specific capacity of LiFePO₄ (about 170mAh g^{-1}).³⁶ During high temperature, the passivation film may crack because of the deformation of metallic lithium. Then fresh Lithium expose to the electrolyte again. Another reason contribute to the capacity fade may be

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the poor interface connection between electrode and electrolyte. Searching for an appropriate anode material or lithium salt by more temperature stable ones maybe a potential solution to this phenomenon. Such as replacing metallic lithium with a lithium alloy and substituting Lithium Bis(trifluoromethanesulfonyl)imide (LiTFSI) or lithium bis(oxalate)borate (LiBOB) for lithium perchlorate.^{7, 37} What's more, interposing an additional thin film of an ion-conducting and electron-insulating oxide between electrode and electrolyte may help decrease the impedance and improving compatibility of interface of interface, as a result to release the capacity fade.³⁸

4. Conclusions

All solid state polymer electrolytes that can fulfill the requirements of safe, flexible have been successfully fabricated by combing branched polymer with electrospun matrix. Among which, Composite electrolytes based on PET possess better morphology, low shrinkage and the highest ionic conductivity. 3PEG@PET electrolytes show good thermal $(250 \degree C)$, electrochemical stability $(4.9V)$ and high temperature discharge capacity $(137.6 \text{ mA} \text{h} \text{g}^{-1})$ at 0.1C. With liquid free and reversible charge/discharge reactions, the elastomer filled electrospun nanofibrous membrane can be a candidate for high performance safe lithium-ion battery.

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Scheme1. Preparation process of foldable electrolyte membranes.

Fig.1. Digital picture of (A) 3PEG and (B) 3PEG@PET electrospun membrane.

Fig.2. SEM images of (A) P(VdF-HFP), (B) 3PEG/PHP, (C) PET, (D) 3PEG@PHP, (E) 3PEG@3PEG/PHP and (F) 3PEG@PET.

Fig.3. Photographs of composite electrolytes (A) before and (B) after uploaded with

3PEG.

Fig.4. (A) TGA curves of composite electrolytes with heating rate of 15 °C min⁻¹, (B) DSC thermograms of 3PEG and electrolytes record from second heating curves with heating rate of 10 $^{\circ}$ C min⁻¹.

Fig.5. Ionic conductivity of 3PEG@3PEG/PHP and 3PEG@PET dependent on thickness of electrospun membranes. Inset shows tensile tests of 3PEG@3PEG/PHP and 3PEG@PET.

Fig.6. Temperature dependence of (A) ionic conductivity of composite electrolytes, (B) their VTF regression curves and (C) ionic conductance curves.

Fig.7. Impedance spectra of 3PEG@PET at different temperature.

Fig.8. Linear sweep voltammetry curves of 3PEG/@3PEG/PHP and 3PEG @PET (scan rate: 5 mV s^{-1}).

Fig.9. (A) second charge/discharge curves at 80 °C and 100 °C and (B) Cycling properties of Li/3PEG@PET/LiFePO₄ at 100 °C. (0.1C rate, 2.5-4V)

Electrolytes	Thickne $ss \,(\mu m)$	σ (S cm ⁻¹) at 30° C	σ (S cm ⁻¹) at 100° C	$G(S)$ at 30 °C	$G(S)$ at 100 °C	$T_{\rm g}$ (°C)	$E_{\rm a}$ (eV)
3PEG	100	1.1×10^{-4}	1.2×10^{-3}	0.021	0.23	-40.65	0.072
3PEG@PHP	30	8.9×10^{-6}	7.8×10^{-5}	0.0059	0.052	-22.44	0.056
3PEG@3PEG/ PHP	30	2.3×10^{-5}	2.4×10^{-4}	0.016	0.16	-27.66	0.058
3PEG@PET	30	4.8×10^{-5}	5.9×10^{-4}	0.032	0.39	-32.06	0.064

Table1. Ionic conductivity, T_g and activation energies of electrolytes.

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Elastomers Uploaded Electrospun Nanofibrous Membrane as Solid State Polymer Electrolytes for Lithium-ion Batteries

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Good electrochemical properties and reversible charge/discharge solid state composite electrolytes achieving in high temperature by uploading 3PEG on electrospun membranes.