A capsule-type gelled polymer electrolyte for rechargeable lithium batteries

W.J. Lia, C.L. Yanga, Z.H. Li*a, Q.Z. Xiaoa, G.T. Leia, Y.H. Dingb

^a Key Laboratory of Environmentally Friendly Chemistry and Applications of Ministry of Education, College of Chemistry, Xiangtan University, Hunan 411105, PR China

^b Institute of Fundamental Mechanics and Materials Engineering, Xiangtan University,

Hunan 411105, PR China

Differential scanning calorimetry (DSC) measurements were performed on a calorimeter (DSC Q10, PerkinElmer, USA) at 5 °C min⁻¹ under a nitrogen atmosphere. The crystallinity (X_c) was calculated using Eq. (1) and the melting enthalpy (ΔH_f) obtained from the DSC curves:

$$X_{\rm c} = \frac{\Delta H_{\rm f}}{\Delta H_{100}} \times 100\% \qquad (1)$$

where ΔH_{100} is the crystalline melting enthalpy of perfectly crystalline PVDF (104.7 J g⁻¹).^{S1}



Figure S1. DSC curves of the trilayer and blended fibrous membranes.

The mechanical properties of the fibrous membranes were investigated using a tensile tester (RG2000-100, Shenzhen Reger Instruments Co., Ltd., China) with rectangular specimens (length: 100 mm; width: 10 mm; thickness: \sim 30 µm). All tests were performed at 28 °C with a speed of 1 mm min⁻¹.

Figure S2 presents the relationship of the breaking elongation and tensile strength of the membranes. It is found that the elongation at break and the maximum stress are 72.6%, and 6.4 MPa for the trilayer fibrous membrane, respectively. The corresponding values are 81.4% and 8.4 MPa for the blended fibrous membrane. It is clear the mechanical performance of the blended fibrous membrane was improved.



Figure S2. Stress-strain curves of the fibrous membranes

Membrane	Electrolyte component	Electrolyte uptake (%)	Ionic conductivity (S cm ⁻¹)	Rate capability (Discharge capacity at various rates) (mAh g ⁻¹)	Cyclability (Capacity retention after n cycles) (%)	Ref.
PAN/PMMA fibers	1M LiTFSI in PYR ₁₄ TFSI+PEGDME	480%	3.6 × 10 ⁻³	139 (0.1C) 134 (0.2C) 120 (0.5C) 101 (1C)	92% (0.2 C, 50 cycles)	S2
P(VdF-HFP) fibers	0.5 M LiTFSI in BMITFSI	750%	2.3×10^{-3}	149 (0.1C) 132 (0.5C)	No data	S3
P(VdF-co-HFP) fibers	1 M LiTFSI in EMImTFSI	700%	4.5×10^{-3}	140 (0.1C)	95% (0.1 C, 25 cycles)	S4
P(VdF-HFP) fibers	0.5M LiTFSI in EMITFSI	No data	9.9×10^{-3}	164 (0.1C)	98.6% (0.1 C, 50 cycles)	S5
BaTiO ₃ /P(VdF-HFP) ibers	0.5 M LiTFSI in BMITFSI	750%	5.2×10^{-3}	165.8 (0.1C)	98.6% (0.1 C, 20 cycles)	S6
P(VdF-HFP) fibers	1M LiTFSI in PY ₁₄ TFSI	No data	<1.0× 10 ⁻³	143 (0.1C) 115 (1C)	92% (0.1 C, 55 cycles)	S7
PVDF-HFP fibers	PMIMTFSI/LiTFSI	No data	1.2×10^{-3}	151.6 (0.1 C)	96.2% (0.1 C,50 cycles)	S 8
SiO ₂ /PVDF-HFP fibers	0.5 M LiTFSI in BMITFSI	400%	4.3×10^{-3}	169 (0.1 C)	94.4% (0.1 C,80 cycles)	S9
CL-PMMA blended PVDF/L-PMMA/PVDF fibers	0.8M LiTFSI in EMITFSI	296%	1.18 ×10 ⁻³	151 (0.1C) 138(0.2C) 124 (0.5C) 110 (1C) 102 (2C)	97% (0.1C, 50 cycles)	This work

Table S1. Comparison of ionic conductivity, rate capability and cyclability of the various ionic liquid-based polymer electrolyte

References

- S1 Nakagawa K.; Ishida Y. J. Polym. Sci. Poly. Phys. 1973, 11, 2153.
- S2 M. Rao, X. Y. Geng, Y. H. Liao, S. J. Hu, W. S. Li, J. Membrane Sci., 2012, 400, 37.
- S3 G. Cheruvally, J. K. Kim, J. W. Choi, et al, J. Power Sources, 2007, 172, 863.
- S4 P. Raghavan, X. Zhao, H. Choi, et al, Solid State Ionics, 2014, 262, 77.
- S5 J. K. Kim, J. Manuel, G. S. Chauhan, et al, *Electrochim. Acta*, 2010, 55, 1366.
- S6 P. Raghavan, X. Zhao, J. Manuel, et al, *Electrochim. Acta*, 2010, 55, 1347.
- S7 J. K. Kim, L. Niedzicki, J. Scheers, et al, J. Power Sources, 2013, 224, 93.
- S8 J. K. Kim, A. Matic, J. H. Ahn, et al, J. Power Sources, 2010, 195, 7639.
- S9 J. K. Kim, G. Cheruvally, X. Li, et al, J. Power Sources, 2008, 178, 815.



Gif. format images of the CGPE (a), TGPE (b) and Celgard porous membrane after soaked in the LiTFSI/EMITFSI electrolyte (c) during streched.

From the images, we can see that during stretched, the CGPE membrane appeared elastic, i.e. it was elongated without rupture; while the TGPE membrane was broken. Obviously, the Celgard porous membrane showed poor wetting capability when it was immersed in the electrolyte of LiTFSI/EMITFSI in spite of good mechanical strength.