A capsule-type gelled polymer electrolyte for rechargeable lithium batteries

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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 \((\Delta H_f)\) obtained from the DSC curves:

\[
X_c = \frac{\Delta H_f}{\Delta H_{100}} \times 100\% \quad (1)
\]

where \(\Delta H_{100}\) is the crystalline melting enthalpy of perfectly crystalline PVDF (104.7 J g\(^{-1}\)).\(^1\)

\[\text{Figure S1. DSC curves of the trilayer and blended fibrous membranes.}\]
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: ~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.

![Stress-strain curves of the fibrous membranes](image)

**Figure S2.** Stress-strain curves of the fibrous membranes
Table S1. Comparison of ionic conductivity, rate capability and cyclability of the various ionic liquid-based polymer electrolyte membranes

<table>
<thead>
<tr>
<th>Membrane</th>
<th>Electrolyte component</th>
<th>Electrolyte uptake (%)</th>
<th>Ionic conductivity (S cm(^{-1}))</th>
<th>Rate capability (Discharge capacity at various rates) (mAh g(^{-1}))</th>
<th>Cyclability (Capacity retention after n cycles) (%)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>PAN/PMMA fibers</td>
<td>1M LiTFSI in PYR(_{14})TFSI+PEGDME</td>
<td>480%</td>
<td>3.6 × 10(^{-3})</td>
<td>139 (0.1C) 134 (0.2C) 120 (0.5C) 101 (1C)</td>
<td>92% (0.2 C, 50 cycles)</td>
<td>S2</td>
</tr>
<tr>
<td>P(VdF-HFP) fibers</td>
<td>0.5 M LiTFSI in BMITFSI</td>
<td>750%</td>
<td>2.3 × 10(^{-3})</td>
<td>149 (0.1C) 132 (0.5C)</td>
<td>No data</td>
<td>S3</td>
</tr>
<tr>
<td>P(VdF-co-HFP) fibers</td>
<td>1 M LiTFSI in EMImTFSI</td>
<td>700%</td>
<td>4.5 × 10(^{-3})</td>
<td>140 (0.1C)</td>
<td>95% (0.1 C, 25 cycles)</td>
<td>S4</td>
</tr>
<tr>
<td>P(VdF-HFP) fibers</td>
<td>0.5 M LiTFSI in EMITFSI</td>
<td>No data</td>
<td>9.9 × 10(^{-3})</td>
<td>164 (0.1C)</td>
<td>98.6% (0.1 C, 50 cycles)</td>
<td>S5</td>
</tr>
<tr>
<td>BaTiO(_3)/P(VdF-HFP) fibers</td>
<td>0.5 M LiTFSI in BMITFSI</td>
<td>750%</td>
<td>5.2 × 10(^{-3})</td>
<td>165.8 (0.1C)</td>
<td>98.6% (0.1 C, 20 cycles)</td>
<td>S6</td>
</tr>
<tr>
<td>P(VdF-HFP) fibers</td>
<td>1 M LiTFSI in PY(_{14})TFSI</td>
<td>No data</td>
<td>&lt;1.0 × 10(^{-3})</td>
<td>143 (0.1C) 115 (1C)</td>
<td>92% (0.1 C, 55 cycles)</td>
<td>S7</td>
</tr>
<tr>
<td>PVDF-HFP fibers</td>
<td>PMIMTFSI/LiTFSI</td>
<td>No data</td>
<td>1.2 × 10(^{-3})</td>
<td>151.6 (0.1 C)</td>
<td>96.2% (0.1 C, 50 cycles)</td>
<td>S8</td>
</tr>
<tr>
<td>SiO(_2)/PVDF-HFP fibers</td>
<td>0.5 M LiTFSI in BMITFSI</td>
<td>400%</td>
<td>4.3 × 10(^{-3})</td>
<td>169 (0.1 C)</td>
<td>94.4% (0.1 C, 80 cycles)</td>
<td>S9</td>
</tr>
<tr>
<td>CL-PMMA blended PVDF/L-PMMA/PVDF</td>
<td>0.8 M LiTFSI in EMITFSI</td>
<td>296%</td>
<td>1.18 × 10(^{-3})</td>
<td>151 (0.1C) 138 (0.2C) 124 (0.5C) 110 (1C) 102 (2C)</td>
<td>97% (0.1C, 50 cycles)</td>
<td>This work</td>
</tr>
</tbody>
</table>
References

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

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.