Supporting Information

Deformable and Flexible Electrospun Supported Cross-linked Gel Polymer Electrolytes for High Safety Lithium Ion Batteries

Yongfen Tong*a, Yuzhong Xu*a, Dian Chen*a, Yu Xie*a, Lie Chenb, Mingming Queb, Yang Houc,d*

aSchool of Environmental and Chemical Engineering, Nanchang Hangkong University, 696 Fenghe South Avenue, Nanchang 330063, China
bCollege of Chemistry, Nanchang University, 999 Xuefu Avenue, Nanchang 330031, China
cDepartment of Chemistry and Food Chemistry & Center for Advancing Electronics, Dresden (CFAED), Technische Universitaet Dresden, 01062 Dresden, Germany
dCollege of Chemical and Biological Engineering, Zhejiang University, Hangzhou 310027, P. R. China
Table S1. Cross-linking agent composition and ionic conductivity (25 °C) of the composite gel polymer electrolytes.

<table>
<thead>
<tr>
<th>Code</th>
<th>Pentaerythritol tetraacrylate (PTA)</th>
<th>Poly(ethylene glycol) diacylate (PEGDA)</th>
<th>Poly(ethylene glycol) methyl ether methacrylate (PEGMA)</th>
<th>Conductivity (S/cm)(^a)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CGPE-1</td>
<td>20</td>
<td>80</td>
<td>0</td>
<td>9.9×10^{-4}</td>
</tr>
<tr>
<td>CGPE-2</td>
<td>20</td>
<td>60</td>
<td>20</td>
<td>1.4×10^{-3}</td>
</tr>
<tr>
<td>CGPE-3</td>
<td>20</td>
<td>50</td>
<td>30</td>
<td>1.7×10^{-3}</td>
</tr>
<tr>
<td>CGPE-4</td>
<td>20</td>
<td>40</td>
<td>40</td>
<td>1.2×10^{-3}</td>
</tr>
<tr>
<td>CGPE-5</td>
<td>20</td>
<td>30</td>
<td>50</td>
<td>6.4×10^{-4}</td>
</tr>
</tbody>
</table>

\(^a\) Ionic conductivity of CGPE with 1.0 M LiTFSI + BMITFSI.
Table S2. Mechanical properties of the composite gel polymer electrolytes.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Stress (MPa)</th>
<th>Modulus (MPa)</th>
<th>Strain (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CGPE-1</td>
<td>6.5</td>
<td>54.0</td>
<td>125.6</td>
</tr>
<tr>
<td>CGPE-2</td>
<td>6.7</td>
<td>58.3</td>
<td>126.8</td>
</tr>
<tr>
<td>CGPE-3</td>
<td>6.8</td>
<td>62.9</td>
<td>128.4</td>
</tr>
<tr>
<td>CGPE-4</td>
<td>6.8</td>
<td>63.8</td>
<td>130.5</td>
</tr>
<tr>
<td>CGPE-5</td>
<td>6.9</td>
<td>72.5</td>
<td>131.7</td>
</tr>
</tbody>
</table>

The mechanical properties of the composite gel polymer electrolytes are presented in Table S1. The value of modulus and strain for the composite membranes increased with the addition of the cross-linking agent and PEGMA. It was suggested that the cross-linked agent enhanced the tensile strength and elastic modulus of electrospun PVDF-HFP, which shows acceptable mechanical properties to be applied in the practice.

Table S3. Ionic conductivity of the composite polymer electrolytes base on different ionic liquid.

<table>
<thead>
<tr>
<th>Code</th>
<th>Pentaerythritol-tetraacrylate</th>
<th>Poly(ethylene)glycol</th>
<th>Poly(ethylene)glycol methyl ether</th>
<th>Ionic liquid</th>
<th>Conductivity (S/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CGPE-BMIBF₄</td>
<td>20</td>
<td>50</td>
<td>30</td>
<td>BMIBF₄</td>
<td>1.9×10⁻³a</td>
</tr>
<tr>
<td>CGPE-3</td>
<td>20</td>
<td>40</td>
<td>40</td>
<td>BMITFSI</td>
<td>1.7×10⁻³b</td>
</tr>
<tr>
<td>CGPE-BMITf</td>
<td>20</td>
<td>30</td>
<td>50</td>
<td>BMITf</td>
<td>8.1×10⁻⁴c</td>
</tr>
</tbody>
</table>

a Ionic conductivity of CGPE with 1.0 M LiBF₄ + BMIBF₄.

b Ionic conductivity of CGPE with 1.0 M LiTFSI + BMITFSI.

c Ionic conductivity of CGPE with 1.0 M LiTf + BMITf.
Figure S1. Variation of ionic conductivity with temperature of CGPE based on electrospun P(VDF-HFP) and cross-linking matrix with different ionic liquid electrolytes.

The ionic conductivities of the composite gel polymer electrolytes based on ionic liquid with different anion (BMITf and BMIBF$_4$) have been investigated. The results demonstrate that the composite gel polymer electrolytes based on BMIBF$_4$ have the maximum values and the order is CGPE-BMIBF$_4$ > CGPE-3 > CGPE-BMITf in all the temperature. It is related to the anionic structure and anionic donating ability. The order of the molar conductivity ratio of the three anions is BF$_4^-$ > TFSI$^-$ > Tf$_2^-$, which results in the CGPE-BMIBF$_4$ based on BMIBF$_4$ possess high ionic conductivity.
The Li||LiFePO$_4$ cell properties with the composite gel polymer electrolytes base on ionic liquid with different anion have been studied. The cells based on BMITFSI (CGPE-3) consistently delivered the highest capacities. The results demonstrate that the anion structure influenced cell performance significantly and the hydrophilic nature of the BMITFSI afford a strong affinity interaction between cross-linking matrix and ionic liquid. It also verifies that the influence of the hydrophilic of the ionic liquid with the polymer matrix may be a critical deterministic factor in cell performance. In some extent, the electrochemical cell performance correlates more strongly with affinity interaction than conductivity.