Supporting Information

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

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Table S1. Cross-linking agent composition and ionic conductivity (25 °C) of the composite gel polymer electrolytes.

Code	Pentaerythritol tetraacrylate (PTA)	Poly(ethylene glycol) diacrylate (PEGDA)	Poly(ethylene glycol) methyl ether methacrylate (PEGMA)	Conductivity (S/cm)[a]
CGPE-1	20	80	0	9.9×10^{-4}
CGPE-2	20	60	20	1.4×10^{-3}
CGPE-3	20	50	30	1.7×10^{-3}
CGPE-4	20	40	40	1.2×10^{-3}
CGPE-5	20	30	50	6.4×10^{-4}

[a] Ionic conductivity of CGPE with 1.0 M LiTFSI + BMITFSI.

Table S2. Mechanical properties of the composite gel polymer electrolyetes.

Samples	Stress(MPa)	Modulus (MPa)	Strain (%)
CGPE-1	6.5	54.0	125.6
CGPE-2	6.7	58.3	126.8
CGPE-3	6.8	62.9	128.4
CGPE-4	6.8	63.8	130.5
CGPE-5	6.9	72.5	131.7

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.

Code	Pentaerythri -tol tetraacrylate (PTA)	Poly(ethylene glycol) diacrylate (PEGDA)	Poly(ethylene glycol) methyl ether methacrylate (PEGMA)	Ionic liquid	Conductivi -ty (S/cm)
CGPE-$BMIBF_4$	20	50	30	$BMIBF_4$	1.9×10^{-3}[a]
CGPE-3	20	40	40	BMITFSI	1.7×10^{-3}[b]
CGPE-BMITf	20	30	50	BMITf	8.1×10^{-4}[c]

[a]Ionic conductivity of CGPE with 1.0 M $LiBF_4$ + $BMIBF_4$.

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

[c]Ionic conductivity of CGPE with 1.0 M LiTf + BMITf.

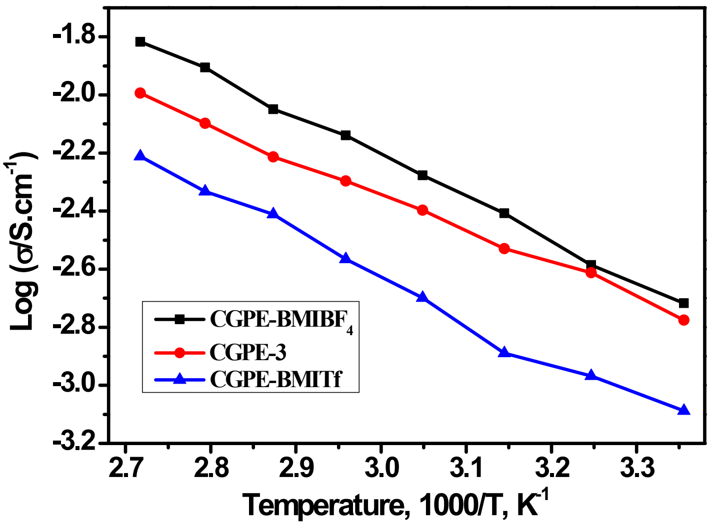

Figure S1. Variation of ionic conductivity with temperature of CGPE based on electrospun P(VDF-HFP) and cross-linking matrix with different ionic liquid electrolytes.

The ionic conductivities of the composite gel polymer electrolytes base on ionic liquid with different anion (BMITf and $BMIBF_4$) have been investigated. The results demonstrate that the composite gel polymer electrolytes base 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^-, which results in the CGPE-$BMIBF_4$ base on $BMIBF_4$ possess high ionic conductivity.

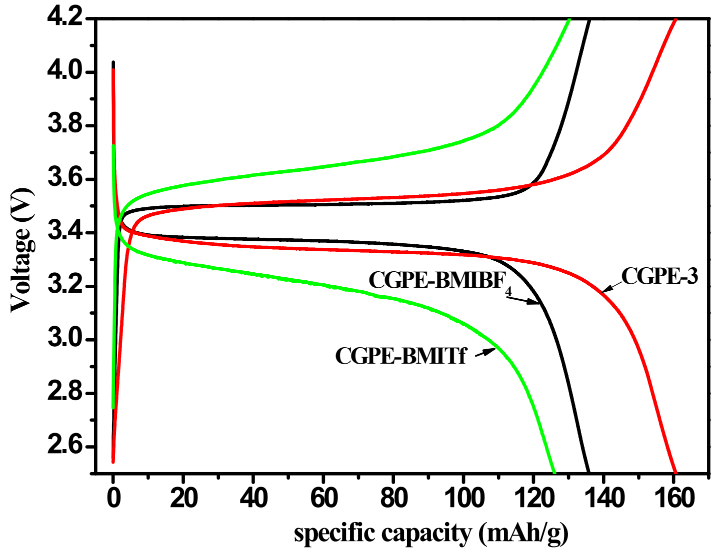

Figure S2. Initial discharge capacities of CGPE based on electrospun P(VDF-HFP)and cross-linking matrix with different ionic liquid electrolytes(Li/PE/$LiFePO_4$ cells, 0.1 C rate, 25 °C).

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.