## **Supporting information**

## Decoupling of Mechanical Strength and Ionic Conductivity in Zwitterionic Elastomer Gel Electrolyte towards Safe Batteries

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## This material includes

Figure S1. Molecular structures of PVMGE, VIPS, MEA, PEGDA and HHMP

Figure S2. Raman spectra characterizing the dissociation effect of PVIPS on LiTFSI

Figure S3. (a) Images of PV<sub>4</sub>M<sub>3</sub>GE before and after the compression; (b) exhibition

of the brittleness of PVIPS gel electrolyte

Figure S4. Frequency dependencies of the storage (G') and loss (G'') moduli of  $PV_1M_1GE$ 

**Figure S5**. Tensile test of the  $PV_1M_1GE$ 

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**Figure S8**. Li plating/stripping voltage profiles of lithium symmetric cells with liquid electrolyte at a current density of 1 mA cm<sup>-2</sup>

Figure S9. Cycling stability of lithium symmetric cell with liquid electrolyte at a current density of  $3 \text{ mA cm}^{-2}$ 

**Figure S10**. Cycling stability of PMEA electrolyte for lithium symmetrical battery at current density of 1 mA cm<sup>-2</sup>

**Figure S11.** SEM image of lithium anode of lithium symmetric battery with liquid electrolyte after cycling for 200 h at current density 1 mA cm<sup>-2</sup>

Figure S12. SEM images of lithium anode of lithium symmetric cell with PMEA after cycling for 200 h at current density 1 mA cm<sup>-2</sup>

**Figure S13**. (a) LSV curve of lithium symmetric cell with liquid electrolyte at a scan rate of 10 mV s<sup>-1</sup>. (b) CV profiles of LiFePO<sub>4</sub>/PV<sub>1</sub>M<sub>1</sub>GE/Li at a scan rate of 0.1 mV s<sup>-1</sup> in a potential window from 2 to 4V. (c) rate performances of LiFePO<sub>4</sub>/Li with liquid electrolyte at 0.5, 1, 2, 3, 5 C; (d) cycling performances of liquid electrolyte at 5 C

Figure S14. Cycle stability of LiFePO<sub>4</sub>/Li cell with PMEA at 5 C

**Table S1**. Comparison of electrochemical performance for  $PV_1M_1GE$  with other electrolytes

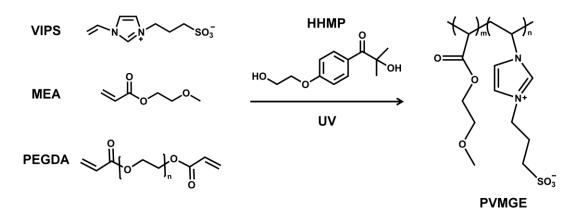
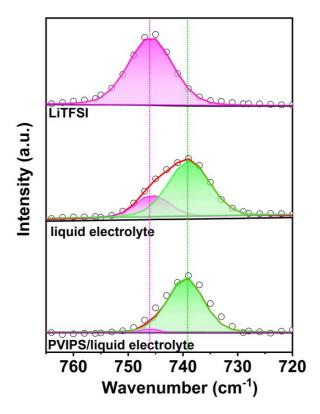


Figure S1. Molecular structures of PVMGE, VIPS, MEA, PEGDA and HHMP.



**Figure S2**. Raman spectra characterizing the dissociation effect of PVIPS on LiTFSI. The band at 739 cm<sup>-1</sup> is related to uncoordinated TFSI<sup>-</sup> while the band at 746 cm<sup>-1</sup> is related to bonded ion pairs. The ratio of uncoordinated TFSI<sup>-</sup> anions and bonded ion pairs is 0.76 in liquid electrolyte and 0.93 in PVIPS/liquid electrolyte, respectively.

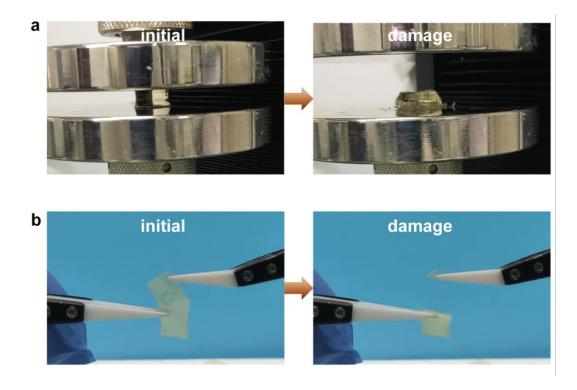


Figure S3. (a) Images of  $PV_4M_3GE$  before and after the compression; (b) exhibition of the brittleness of PVIPS gel electrolyte.

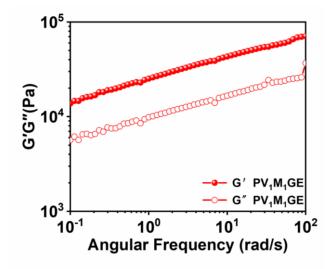


Figure S4. Frequency dependencies of the storage (G') and loss (G'') moduli of  $PV_1M_1GE$ .

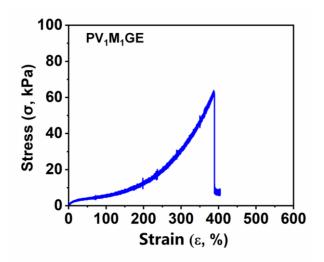


Figure S5. Tensile test of the  $PV_1M_1GE$ .

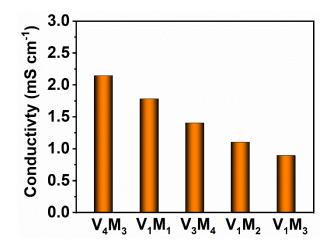


Figure S6. Ionic conductivity of the PVMGEs with different VIPS/MEA weight ratios.

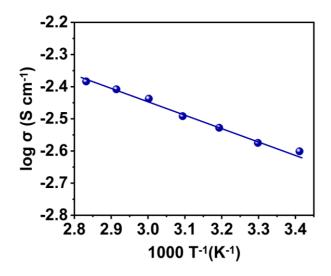
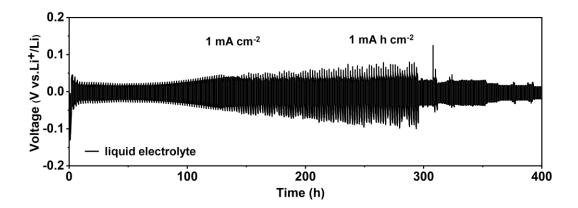
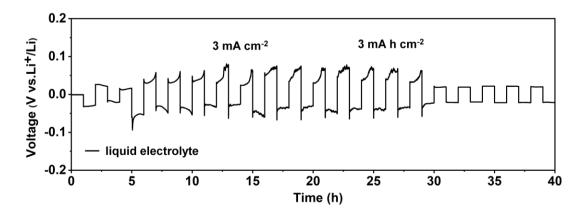


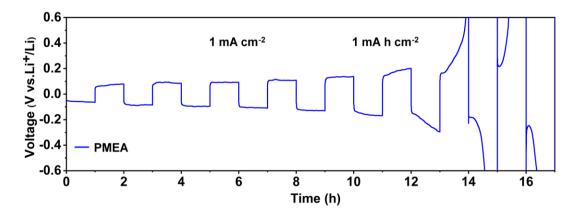
Figure S7. Temperature dependence of ionic conductivity of PV<sub>1</sub>M<sub>1</sub>GE.



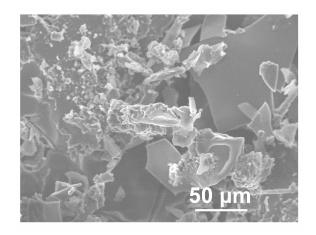
**Figure S8**. Li plating/stripping voltage profiles of lithium symmetric cells with liquid electrolyte at a current density of  $1 \text{ mA cm}^{-2}$ .



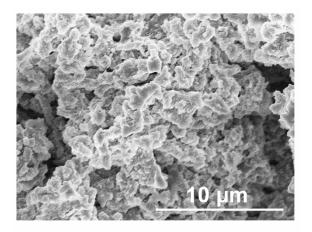
**Figure S9**. Cycling stability of lithium symmetric cell with liquid electrolyte at a current density of 3 mA cm<sup>-2</sup>.



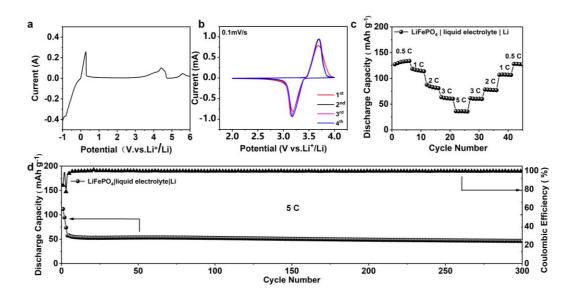
**Figure S10**. Cycling stability of PMEA electrolyte for lithium symmetrical battery at current density of 1 mA cm<sup>-2</sup>.



**Figure S11**. SEM image of lithium anode of lithium symmetric battery with liquid electrolyte after cycling for 200 h at current density 1 mA cm<sup>-2</sup>.



**Figure S12**. SEM images of lithium anode of Li/PMEA/Li cell after cycling for 200 h at current density 1 mA cm<sup>-2</sup>.



**Figure S13**. (a) LSV curve of lithium symmetric cell with liquid electrolyte at a scan rate of 10 mV s<sup>-1</sup>. (b) CV profiles of LiFePO<sub>4</sub>/PV<sub>1</sub>M<sub>1</sub>GE/Li at a scan rate of 0.1 mV s<sup>-1</sup> in a potential window from 2 to 4V. (c) rate performances of LiFePO<sub>4</sub>/Li with liquid electrolyte at 0.5, 1, 2, 3, 5 C; (d) cycling performances of liquid electrolyte at 5 C.

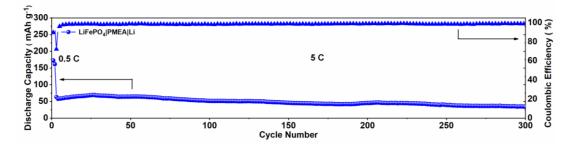


Figure S14. Cycling performance of LiFePO<sub>4</sub>/Li cell with PMEA electrolyte at 5 C.

GPE	Li/Li		LiFePO4/Li		Ref.
	Current density (mA cm <sup>-2</sup> )	Cycle Lifespans (h)	Current density (1 C=170 mA g <sup>-1</sup> )	Discharge capacity (mA h g <sup>-1</sup> )	
$PV_1M_1GE$	1	700	5 C	70	Our work
PVDF- HFP	1	250	2 C	57	1
SiO <sub>2</sub> -GPE	1	400	0.1 C	125	2
glass fiber- poly (butyl acrylate)	0.5	~250	0.5 C	84	3
PAN-PEO	-	-	0.1 C	149	4
cellulose aerogel membranes	-	-	0.5 C	134	5
double polymer network-GPE	0.5	<400	0.5	124	6

**Table S1.** Comparison of electrochemical performance for  $PV_1M_1GE$  with other electrolytes.

## References

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