

## Supporting Information for

### A Facile In Situ Approach to Ion Gel Based Polymer Electrolyte for Flexible Lithium Batteries

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## 1. Experimental Section

### 1.1 Synthesis of imidazolium based monomer MHBIm-TFSI

The synthesis of imidazolium based monomer, 1-[(2-methacryloyloxy) hexyl]-3-butylimidazolium bis(trifluoromethanesulphonyl) imide (MHBIm-Br) as shown in **Scheme S1**. Typically, 19 g (0.105 mol) 6-bromo-1-hexanol in 30 mL of dichloromethane was added to a two-neck 100-mL flask and stirred for 10 min in an ice bath. Then, 11.1 g (0.11 mol) of triethylamine along with 10 mL of dichloromethane was added slowly by a funnel under N<sub>2</sub> atmosphere. After stirring the mixture for 10 min, 10.5 g (0.1 mol) methacryloyl chloride in 10 mL dichloromethane was added dropwise. The reaction mixture was stirred 12 h at room temperature and then filtered. Washing the filtrate with 200 mL of DI water four times and collecting the organic layer. Remaining water was further dried with anhydrous magnesium sulfate. Then dichloromethane in the mixture was removed by vacuum, after which clear liquid 3-bromohexyl methacrylate yielded (70% yield).

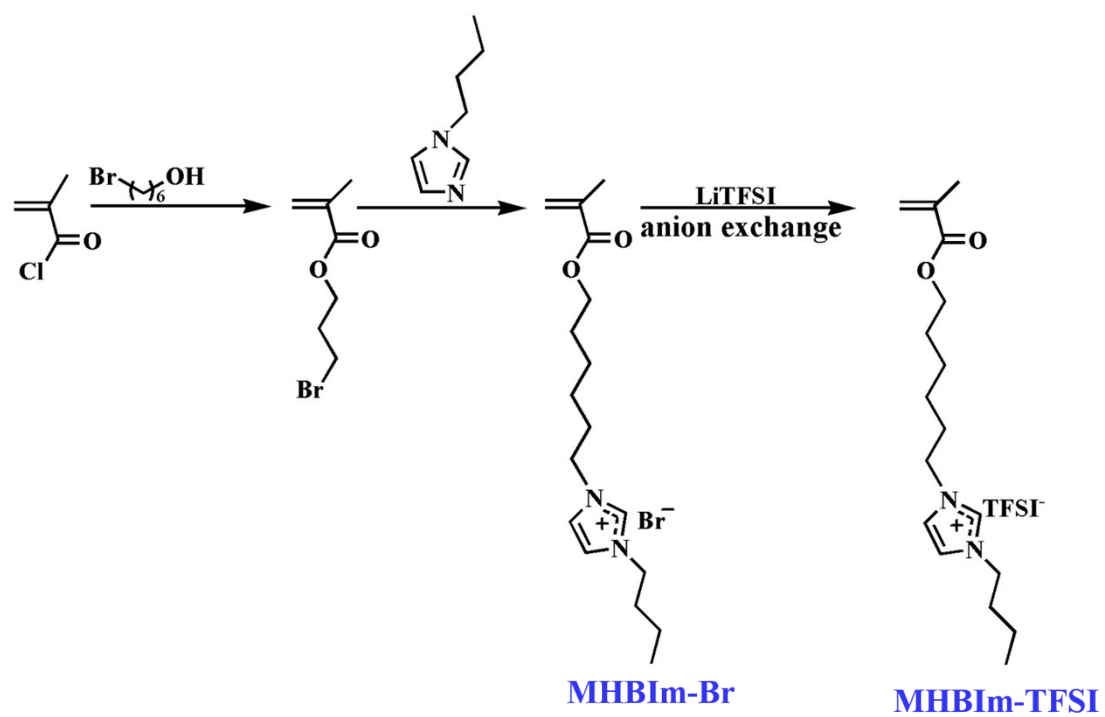
To obtain imidazolium bromide monomer, a mixture consists of 16.3 g (0.07 mol) 3-

bromopropyl methacrylate and 8.69 g (0.07 mol) 1-butyl imidazole and a small amount of 2,6-di-tertbutyl-4-methylphenol (inhibitor), was stirred in a 40 °C oil bath for 24 h, after which viscous liquid was obtained (80% yield). The resulting liquid was dissolved in 30 mL dichloromethane and re-precipitated in 200 mL diethyl ether in an ice bath.

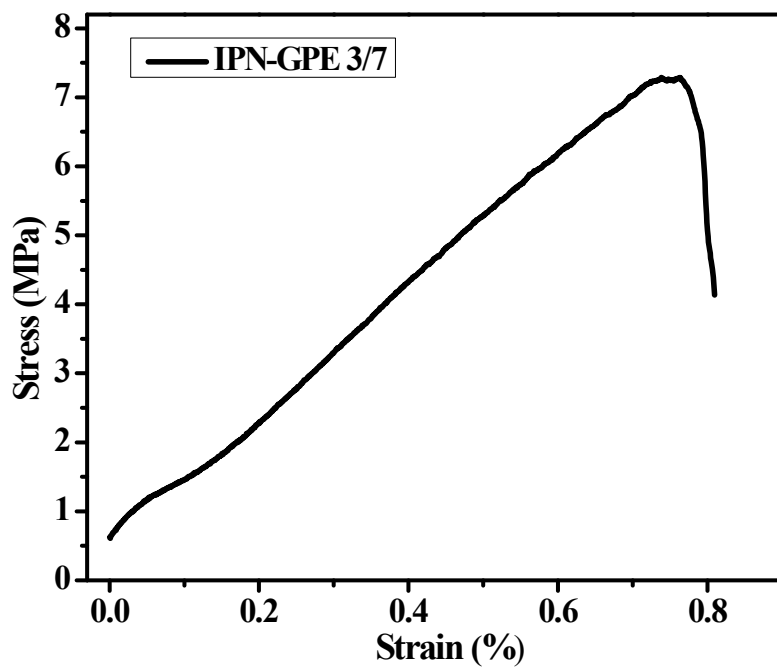
Imidazolium bis(trifluoromethane sulfonyl)imide monomer MHBIm-TFSI is prepared by a simply ion-exchange method. Briefly, LiTFSI is added into homogeneous acetone solution of MHBIm-Br with the anion mole ratio of TFSI/Br<sup>-</sup> =3/1. The raw product was purified via washing with deionized water several times to remove the superfluous lithium salts. The organic layer was further dried with anhydrous magnesium sulfate. Then remained solvents in the mixture were further removed by vacuum. After that pure MHBIm-TFSI was obtained. <sup>1</sup>H NMR (δppm, DMSO-d<sub>6</sub>): <sup>1</sup>H NMR (δppm, DMSO-d<sub>6</sub>): 9.16 (s, 1H, N-CH=N), 7.76 (d, 2H, N-CH-CH-N), 5.98 (s, 1H, HCH=C(CH<sub>3</sub>)), 5.62 (s,1H, HCH=C(CH<sub>3</sub>)), 4.15 (t, 2H, N-CH<sub>2</sub>-(CH<sub>2</sub>)<sub>4</sub>-CH<sub>2</sub>-O), 4.05 (t, 2H, N-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-O) 3.93 (t, 2H, N-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>3</sub>), 1.85 (s, 3H, CH<sub>2</sub>=C(CH<sub>3</sub>)).

## 1.2 preparation of PVDF-HFP electrospun membrane

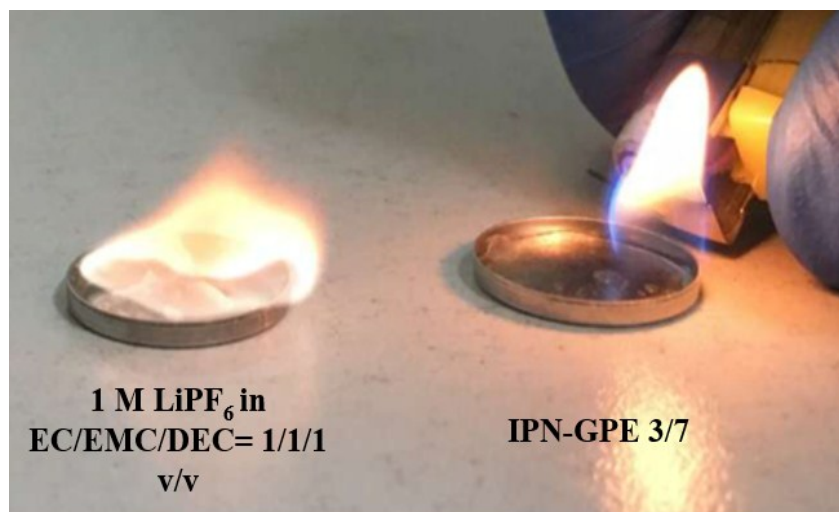
PVDF-HFP nanofibrous membranes are prepared by electrospinning. A 16 wt. % solution of PVDF-HFP for electrospinning was prepared by dissolving PVDF-HFP in a mixture of acetone and N, N-dimethylformamide (DMF) (3/1, V/V) by mechanical stirring over night. The solution was taken in the syringe and delivered with a flow rate of 1.2 ml h<sup>-1</sup>. Electrospinning progress was conducted at an electric voltage of 20 kV at room temperature with the above solution, the distance between target drum and the syringe needle tip (inner diameter 22 μm) was 15 cm.



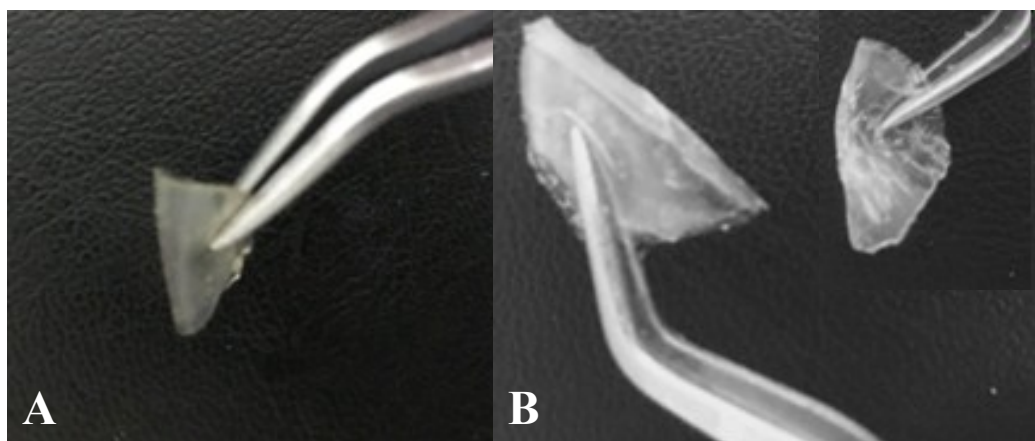
**Scheme S1.** Synthesis route of imidazolium based monomer MHBIIm-TFSI.



**Fig. S1** Stress-strain curve of IPN-GPE 3/7 electrolyte membrane.



**Fig. S2** Combustion testing of 1 M LiPF<sub>6</sub> EC/EMC/DEC liquid electrolyte (left) and IPN-GPE 3/7 (right).



**Fig. S3** Electrolyte membrane prepared with monomers of (A) PEGMA and PEGDA, and (B) MHBIm-TFSI and PEGDA.

**Table S1.** The individual components in IPN-GPE 10, IPN-GPE 8/2, IPN-GPE 5/5, and IPN-GPE 3/7.

| Samples      | MHBI <sub>m</sub> -TFSI<br>(wt%) | PEGDA<br>(wt%) | EMIM-TFSI<br>(wt%) | LiTFSI<br>(wt%) |
|--------------|----------------------------------|----------------|--------------------|-----------------|
| IPN-GPE 10   | 64                               | 20             | 0                  | 16              |
| IPN-GPE 8/2, | 55.7                             | 17.4           | 9.5                | 17.4            |
| IPN-GPE 5/5, | 40                               | 12.7           | 27.3               | 20              |
| IPN-GPE 3/7  | 26.8                             | 8.3            | 42.6               | 22.3            |

**Table S2** Summary of  $T_g$  values for different electrolytes.

| Electrolyte composition<br>(polymer/electrolyte) | Glass transition<br>temperatures ( $T_g/^\circ\text{C}$ ) | Ref.             |
|--|---|------------------|
| 3PEG@PHP   | -22.44  | [1]              |
| 3PEG@3PEG/PHP                                    | -24.66  |                  |
| 3PEG   | -40.65  |                  |
| 3PEG@PET   | -33.06  |                  |
| semi-IPN) SPE                                    | -58   | [2]              |
| 3P(MPBI <sub>m</sub> -Tf)                        | 33.4  | [3]              |
| 3P(MPBI <sub>m</sub> -Br)                        | 71.5  |                  |
| 3P(MPBI <sub>m</sub> -TFSI)                      | 3.3   |                  |
| ipn-PEA  | -64.2   | [4]              |
| ETPTA/EGMEA                                      | 40  | [5]              |
| SPE0   | -55.2   | [6]              |
| SPE3   | -24.8   |                  |
| GPE12  | -62.3   |                  |
| C-PEGDE  | -49.2   | [7]              |
| PEO-SPE  | -25   | [8]              |
| PPC  | 24.4  |                  |
| PPC-SPE  | 4.7   |                  |
| CPPC-SPE   | 4.8   |                  |
| <b>EMIM-TFSI</b>                                 | <b>-86</b>  | <b>This work</b> |
| <b>IPN-GPE 3/7</b>                               | <b>-74</b>  |                  |
| <b>IPN-GPE 5/5</b>                               | <b>-56</b>  |                  |
| <b>IPN-GPE 8/2</b>                               | <b>-34</b>  |                  |
| <b>IPN-GPE 10/0</b>                              | <b>-27</b>  |                  |



**Table S3** Capacity values of lithium ion batteries assembled with different electrolytes.

| Electrolyte composition<br>(polymer/electrolyte) | Capacity (mA h g <sup>-1</sup> )                       | Ref.                 |
|--|--|----------------------|
| OEGMA-BnMA/LiPF <sub>6</sub> -EC-DMC             | 122@0.1 C, 25@5 C                                      | [9]                  |
| EO-based/LiBF <sub>4</sub> -EC-GBL               | 111@1/24 C 80%   | [10]                 |
| EO-based/LiFSI-EC-GBL                            | 156@0.04 C   | [11]                 |
| P(EO-MEEGE)/LiTFSI-AN                            | 127@1/24 C   | [12]                 |
| semi-IPN SPE                                     | 147@0.1 C, 132@0.5 C                                   | [2]                  |
| C-PEGDE  | 135@0.05C, 115@0.1C,<br>100@0.15C, 80@0.2 C            | [7]                  |
| P(EO-PO)/LiPF <sub>6</sub> -EC-DMC-DEC           | 125@0.5 C, 88@5 C,<br>12@17 C                          | [13]                 |
| 3P(MPBIIm-TFSI)                                  | 124@0.5 C, 101@1 C                                     | [3]                  |
| PVCA-SPE   | 124@0.2 C, 73@0.5C                                     | [14]                 |
| ipn-PEA  | 141@0.5 C, 66@5 C                                      | [4]                  |
| <b>IPN-GPE 3/7</b>                               | <b>152.1@0.1 C, 146@0.2 C,<br/>132@0.5 C 107.6@1 C</b> | <b>This<br/>work</b> |

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