## **Supporting Information**

## Facile fabrication of robust gel poly(ionic liquid) electrolytes via bases treatment at room temperature

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> а 1) 200 °C, 6 h High energy consumption 2 CF<sub>3</sub>SO<sub>3</sub>H, r. t., 24 h Corrosive, difficult to remove 3 NH<sub>3</sub>, r. t., 6 h Mild, simple to operate 50 wt% IL GPE-IL-5 b Acetone ③ Room temperature 1 Thermal ②No crosslinking crosslinking crosslinking

**Figure S1.** a) Schematic preparation of GPE-IL-5 via different methods including thermal annealing,  $CF_3SO_3H$  catalysis and ammonia annealing, respectively; b) Photographs of the membranes treated by heat, acid and ammonia, respectively before and after soaking in acetone. Please note:  $CF_3SO_3H$  vaporizes quickly, and dissolves the GPE-IL-5 precursor, yielding no membranes.



Figure S2. <sup>1</sup>H NMR spectrum of CMVImBr monomer.



Figure S3. <sup>1</sup>H NMR spectrum of PCMVImTFSI.



**Figure S4.** a) Photographs of GPE-IL-8 before and after  $NH_3$  vapor treatment, b) storage modulus (G') and loss modulus (G") of GPE-IL-8 before and after  $NH_3$  treatments as a function of sweeping frequency.



**Figure S5.** (a) Stress-strain curves of PCMVImTFSI membranes treated in NH<sub>3</sub> vapor for varied time. (b) Effect of NH<sub>3</sub> annealing time on fracture strength and elongation at break of PCMVImTFSI membranes.



Figure S6. ATR-FTIR spectra of the PCMVImTFSI membranes without any treatment (black line) and with  $NH_3$  treatment for 2 days (red line) and 0.1 M NaOH treatment for 2 days (blue line).



**Figure S7.** (a) Photographs of membranes made from PCMVImX (X = Br, TFSI,  $BF_{4}$ -,  $PF_{6}^{-}$  and  $Bph_{4}^{-}$ ). (b) Photograph of poly(ionic liquid) containing triazolium cation (PCMVTzTFSI) (NH<sub>3</sub> treated). Please note: these membranes are insoluble in DMF or DMSO. (c,d) ATR-FTIR spectra of PCMVImX (X = Br, TFSI<sup>-</sup>,  $BF_{4}^{-}$ ,  $PF_{6}^{-}$  and  $Bph_{4}^{-}$ ) before and after NH<sub>3</sub> treatment. (e) ATR-FTIR spectra of PCMVTzTFSI (the cation is triazolium) before and after NH<sub>3</sub> treatment.



**Figure S8.** (a) Chemical structure of a copolymer ( $P_{co}$ ) containing 20 mol% nitrile monomer. (b) The photograph of  $P_{CO}$  (NH<sub>3</sub> treated) membrane immersed in DMF (note: the membrane is insoluble in DMF). (c) Stress-strain curves of  $P_{co}$  membrane before and after NH<sub>3</sub> treatment.



**Figure S9.** a) Effect of ammonia treatment time on fracture strength and elongation at break of GPE-IL-5; b) Effect of heat treatment time on fracture strength and elongation at break of GPE-IL-5.



**Figure S10.** (a) Actual IL content of GPE-IL-5 with different NH<sub>3</sub> treatment time; (b) Actual IL content of GPE membranes with different IL content (IL is extracted by ethanol, and the actual IL content is calculated by mass change).



**Figure S11.** Stress-strain curves of GPE-IL-5 membrane treated with heat (200 °C) and NH<sub>3</sub> vapor, respectively.



Figure S12. Cyclic tension of GPE-IL-5 with NH<sub>3</sub> treatment at a strain of 10%.



Figure S13. SAXS curves of GPE-IL-X (X = 4-8) (beam size:  $0.8 \times 0.8$  mm<sup>2</sup>; pixel size:  $0.172 \times 0.172$  mm<sup>2</sup>; wavelength: 0.134144 nm; angle of incidence: 0.2 degree).