

Electronic Supplementary Information (ESI) of

## Preparation and characterization of gel polymer electrolytes using poly(ionic liquids) and high lithium salt concentration ionic liquids

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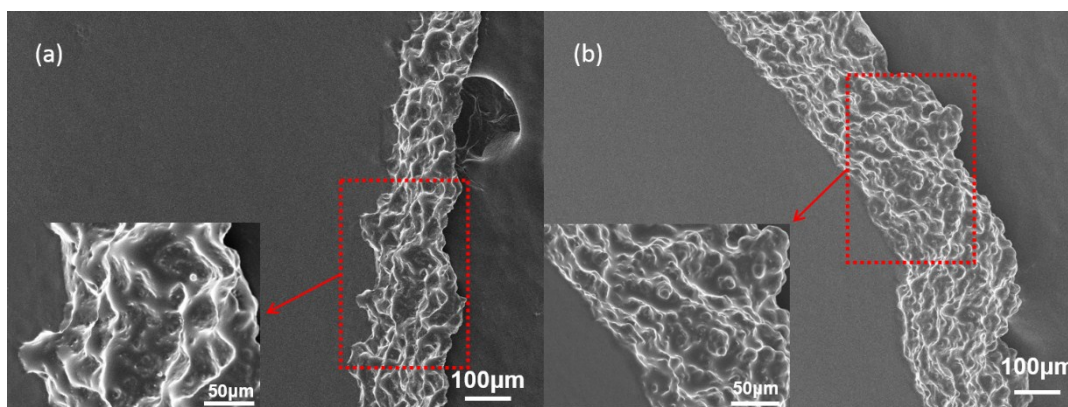
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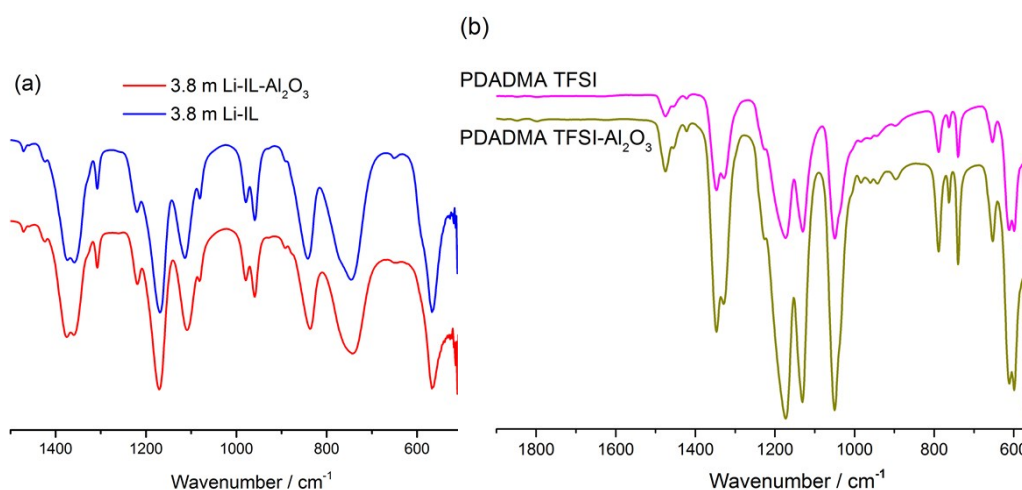
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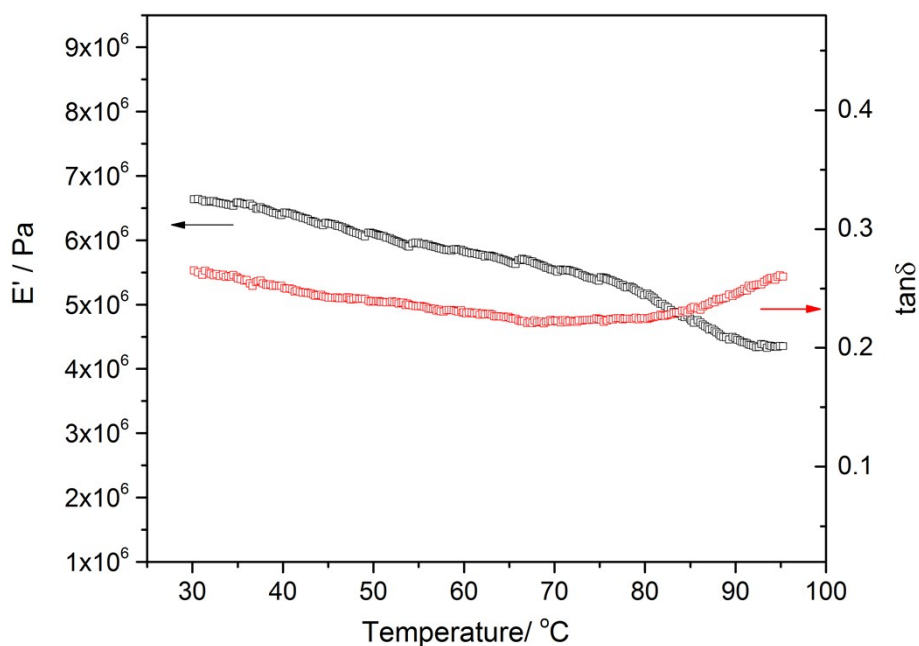


**Figure S1.** The SEM images of (a) GPE-40/60- $\text{Al}_2\text{O}_3$  and (b) GPE-50/50- $\text{Al}_2\text{O}_3$ . The insets are the zoom-in images of highlighted cross-sectional areas.



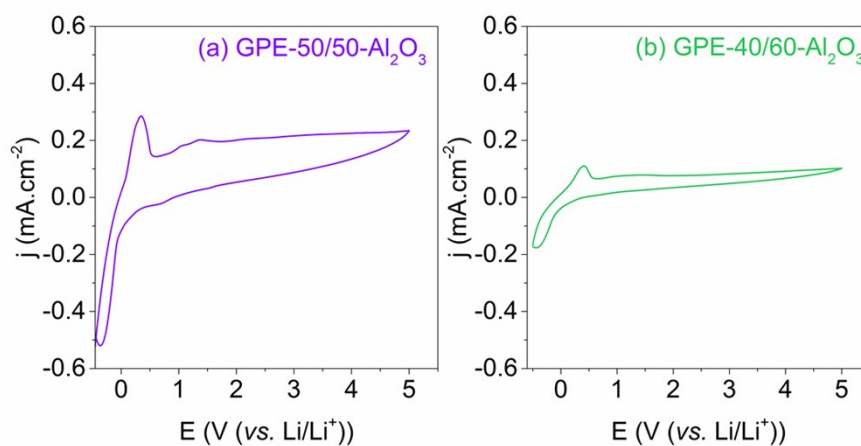
**Figure S2.** The comparison of FTIR spectra for (a) 3.8 m Li-IL samples with and without  $\text{Al}_2\text{O}_3$  nano-particles; (b) PDADMA TFSI samples with and without  $\text{Al}_2\text{O}_3$  nano-particles.

The mechanical properties is critically important for gel electrolytes especially in the applications of all-solid-state batteries. Thus, we selected the GPE-50/50- $\text{Al}_2\text{O}_3$  to investigate the mechanical behaviour by dynamic mechanical analysis (DMA). The DMA measurement was done under compression mode in  $\text{N}_2$ -filled environmental box, the  $\text{H}_2\text{O}$  level was less than 100 ppm. As shown in Figure S3, the storage elastic modulus ( $E'$ ) decreases with increasing of temperature which could result from the thermal relaxation of the polymer component. But it should be noted that the elastic modulus at 30 °C is relatively high, at 6.4 MPa.



**Figure S3.** Temperature dependence of storage elastic modulus ( $E'$ ) and dissipation factor ( $\tan \delta$ ) for GPE-40/60- $\text{Al}_2\text{O}_3$ . The frequency is 1Hz and heating rate is 2 °C/min.

For the high voltage behaviour (up to +5 V vs.  $\text{Li}/\text{Li}^+$ ) we measured linear sweep voltammograms (CV) from -0.5 to +5 V vs.  $\text{Li}/\text{Li}^+$  for the promising GPEs (both GPE-50/50- $\text{Al}_2\text{O}_3$  and GPE-40/60- $\text{Al}_2\text{O}_3$  electrolytes) at a stainless steel (SS) working electrode with a potential sweep rate of 10  $\text{mV s}^{-1}$  at 50 °C. Reversible Li plating/stripping was observed and higher current densities were reached in the case of the GPE-50/50- $\text{Al}_2\text{O}_3$ . The results confirm a high voltage stability of the promising GPE.



**Figure S4.** Linear sweep voltammograms (1st cycle) for GPEs with different composition (GPE-50/50-Al<sub>2</sub>O<sub>3</sub> and GPE-40/60-Al<sub>2</sub>O<sub>3</sub> electrolytes) at a stainless steel (SS) working electrode with a potential sweep rate of 10 mV s<sup>-1</sup> at 50 °C.