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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**Figure S1.** The SEM images of (a) GPE-40/60-Al$_2$O$_3$ and (b) GPE-50/50-Al$_2$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 Al$_2$O$_3$ nano-particles; (b) PDADMA TFSI samples with and without Al$_2$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-Al$_2$O$_3$ to investigate the mechanical behaviour by dynamic mechanical analysis (DMA). The DMA measurement was done under compression mode in N$_2$-filled environmental box, the H$_2$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 δ ) for GPE-40/60-Al2O3. The frequency is 1Hz and heating rate is 2 °C/min.

For the high voltage behaviour (up to +5 V vs. Li/Li+) we measured linear sweep voltammograms (CV) from -0.5 to +5 V vs. Li/Li+ for the promising GPEs (both GPE-50/50-Al2O3 and GPE-40/60-Al2O3 electrolytes) at a stainless steel (SS) working electrode with a potential sweep rate of 10 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-Al2O3. 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-Al₂O₃ and GPE-40/60-Al₂O₃ electrolytes) at a stainless steel (SS) working electrode with a potential sweep rate of 10 mV s⁻¹ at 50 °C.