Supplementary Information for

Inherent limitations of the hydrogen-bonding UPy motif as self-healing functionality for polymer electrolytes

Cuc Thu Mai, Harish Gudla, Guiomar Hernández, Kristina Edström, Jonas Mindemark

Department of Chemistry – Ångström Laboratory, Uppsala University, Uppsala, Sweden

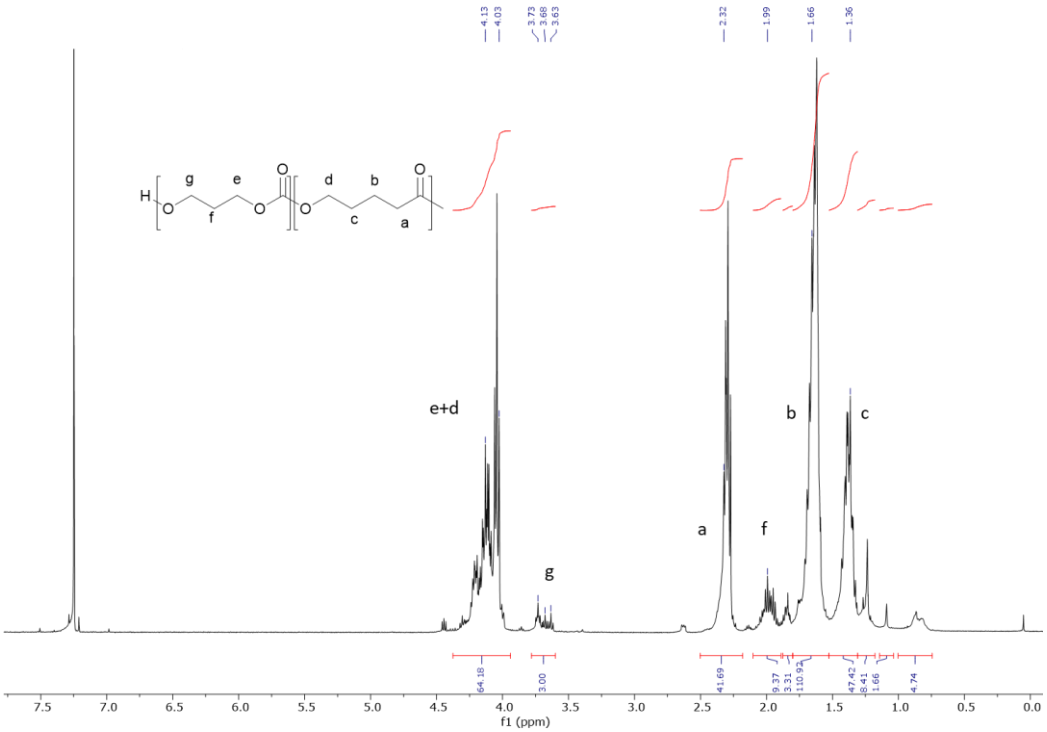

Figure S1. ^{1}H NMR spectrum of star-branched PCL-PTMC.

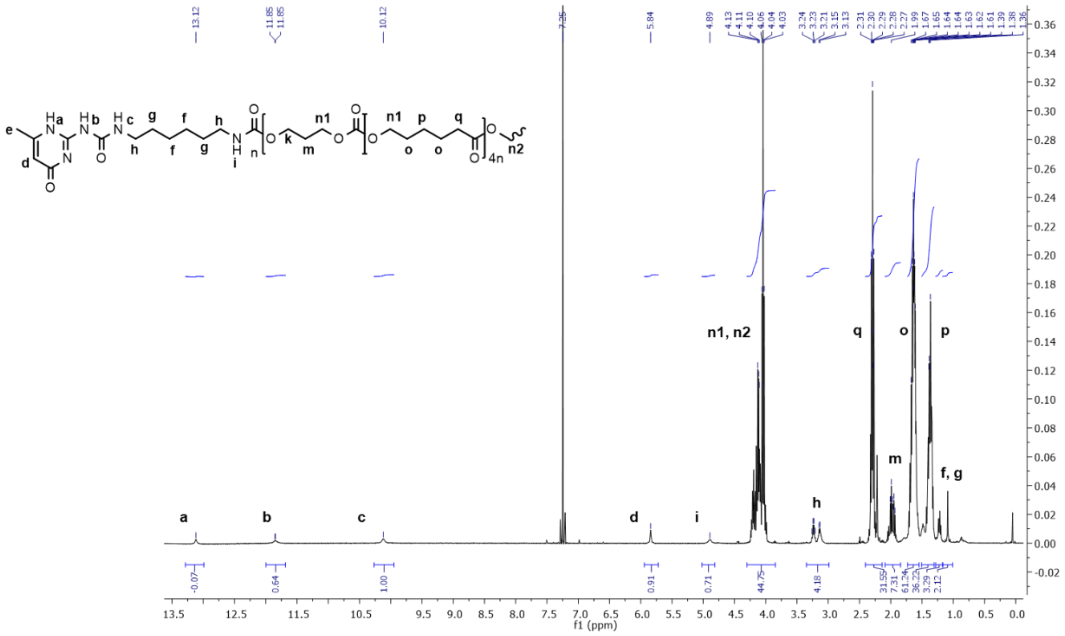

Figure S2. ^{1}H NMR spectrum of shPCL-PTMC.

Table S1. Summary of thermal properties and ionic conductivity of unmodified PCL-PTMC and self-healing polymer electrolytes.

Sample	σ (S cm^{-1}) at 30 °C	T_g (°C)	T_m (°C)
PCL-PTMC	1.2×10^{-5}	−53.3	54.6 & 69.9
shPCL-PTMC + LiTFSI	2.0×10^{-6}	−26.3	-
shPCL-PTMC + $LiPF_6$	1.3×10^{-7}	−17.4	-
shPCL-PTMC + NaTFSI	3.0×10^{-6}	−27.6	48.8 & 72.3

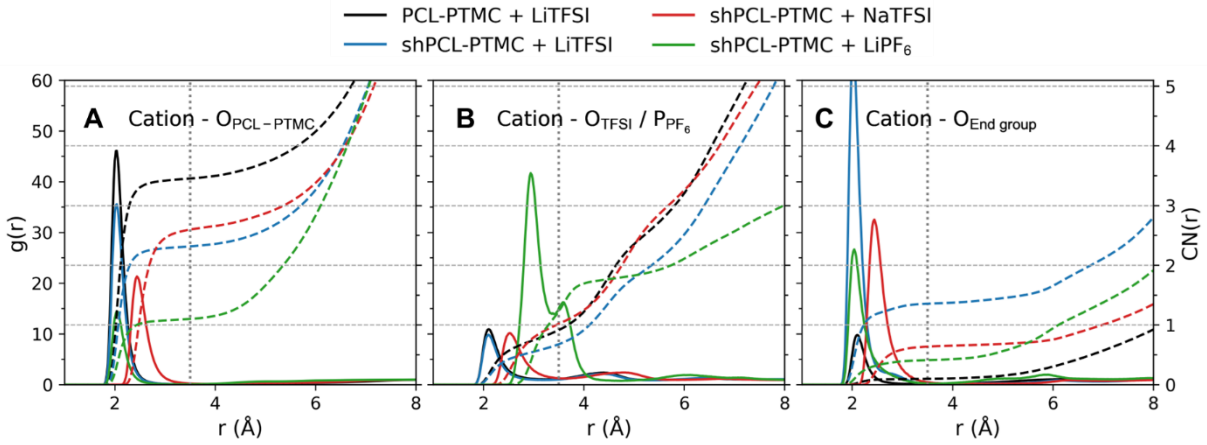

Figure S3. The radial distribution function ($g(r)$) and coordination number (CN(r)) of Li^+/Na^+ with carbonyl O from PCL-PTMC (A), O from $TFSI^-$ or P form PF_6^- (B) and O atoms from end groups i.e., OH for PCL-PTMC and UPy for shPCL-PTMC (C) with salts.

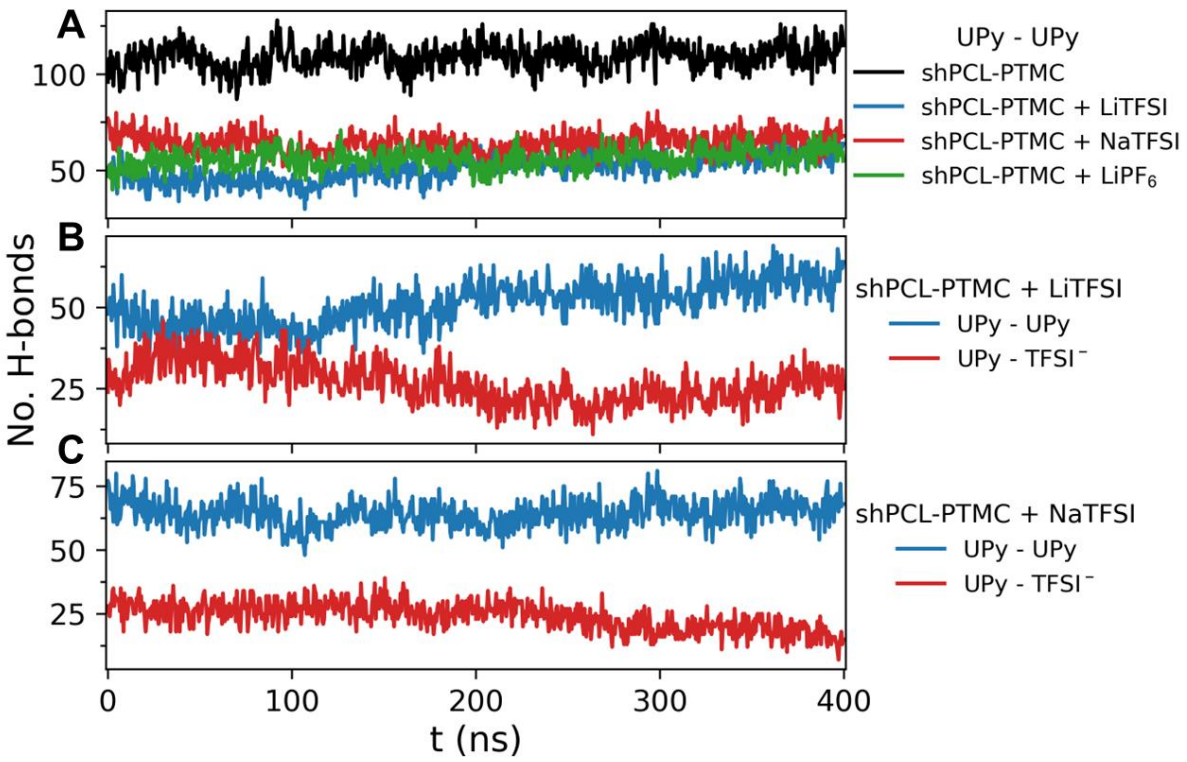

Figure S4. The total number of H-bonds as a function of simulation time between the self-healing groups (UPy – UPy) and between self-healing and $TFSI^-$ groups (UPy – $TFSI^-$) for different polymer electrolyte systems, i.e., shPCL-PTMC with LiTFSI, NaTFSI, and $LiPF_6$.