Electronic Supplementary Information (ESI)

Controlling mechanical properties of ultrahigh molecular weight ion gels by chemical structure of ionic liquids and monomers

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Fig. S1. GPC traces for UHMW polymers, which were extracted from the as prepared UHMW gels.



Fig. S2. ¹H-NMR spectra for as prepared UHMW gels in CDCl₃. Monomer conversions were calculated by comparing the residual vinyl peak with the polymer peaks. The methyl peak for PMMA, the methylene peaks for PEMA and PPMA were used for calculation.



Fig. S3. Dependence of molecular weight of PMMA synthesised by *in situ* radical polymerisation in various ILs on ionic strength of the precursor solutions. The dotted line is a guide to the eye.



Fig. S4. tTS master curves of tan $\delta (= G''/G')$ for UHMW gels with different IL and polymer structures. In each graph, only the master curve of interest is highlighted, while the other curves are shown in grey to clarify their relative positions.



Fig. S5. Frequency sweep measurements of storage (G', closed) and loss (G'', open) moduli for UHMW gels at a temperature of 120 °C with a strain amplitude of 1%. In each graph, only the master curve of interest is highlighted, while the other curves are shown in grey to clarify their relative positions.



Fig. S6. Heat flow curves for the neat $[C_2mIm][TFSI]$ (a) and the UHMW PMMA/ $[C_2mIm][TFSI]$ ion gel (b) at a heating rate of 10 °C min⁻¹.



Fig. S7. Relationship between the glass transition temperature (T_g) and Young's modulus *(E)* of ion gels.



Fig. S8. Comparison of the stress-strain curves for pristine (left) and healed (right) PMMA/[C₂mIm][TFSI] and PMMA/[C₁₂mIm][TFSI] UHMW gels.



Fig. S9. $G_{total}(r)$ functions for the PMMA/[C₂mIm][TFSI], PMMA/[C₁₂mIm][TFSI], and PEMA/[C₂mIm][TFSI] systems.

Table S1. Density d, compositions (number of ion-pairs and polymer molecules), and

System	$d [g m L^{-1}]$		IL	PMMA	PEMA	Dou longth [Å]	
	Calc	MD	(ion pair)	(20mer)	(20mer)	box length [A]	
PMMA/[C ₂ mIm][TFSI]	1.402	1.403	256	20	-	55.45	
PMMA/[C ₁₂ mIm][TFSI]	1.223	1.214	256	34	-	65.50	
PEMA/[C ₂ mIm][TFSI]	1.379	1.373	256	-	18	55.64	

box length of the systems for MD simulations.

Polymer	Salvant	Young's modulus	Fracture	Fracture	$T / \Omega Ch$
	Solvent	(E) / kPa ^a	stress / kPa ^a	strain / %ª	I_{g}/C^{0}
РММА	[C ₁ mIm][TFSI]	147	271	700	-62.1
	[C ₂ mIm][TFSI]	124	190	653	-70.0
	[C ₃ mIm][TFSI]	94	182	799	-68.0
	[C ₈ mIm][TFSI]	120	225	853	-60.3
	[C ₁₂ mIm][TFSI]	101	187	656	-56.1
	[C ₂ dmIm][TFSI]	167	353	578	-62.4
	[C ₂ mIm][BETI]	196	467	454	-57.2
	$[C_2mIm][IM_{14}]$	180	665	625	-69.7
PEMA	[C ₂ mIm][TFSI]	71	106	1711	-78.7
PPMA	[C ₂ mIm][BETI]	69	135	639	-74.5

Table S2. Summary of the tensile properties and glass transition temperature (T_g)

for UHMW gels.

^ameasured by tensile tests. ^bdetermined by DSC measurements.