Tough Polymeric Hydrogels using Ion-Pair Comonomers

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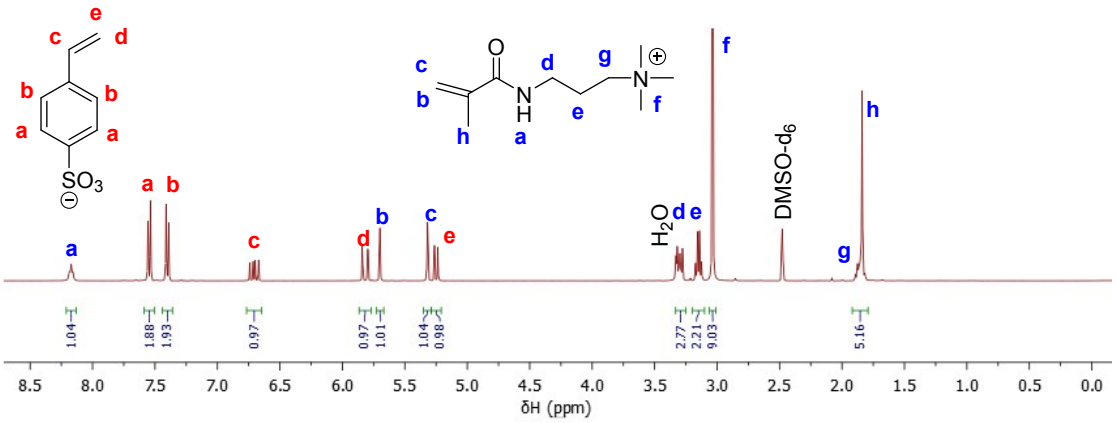

Fig. S1 ^{1}H NMR of the IPC salt **1**

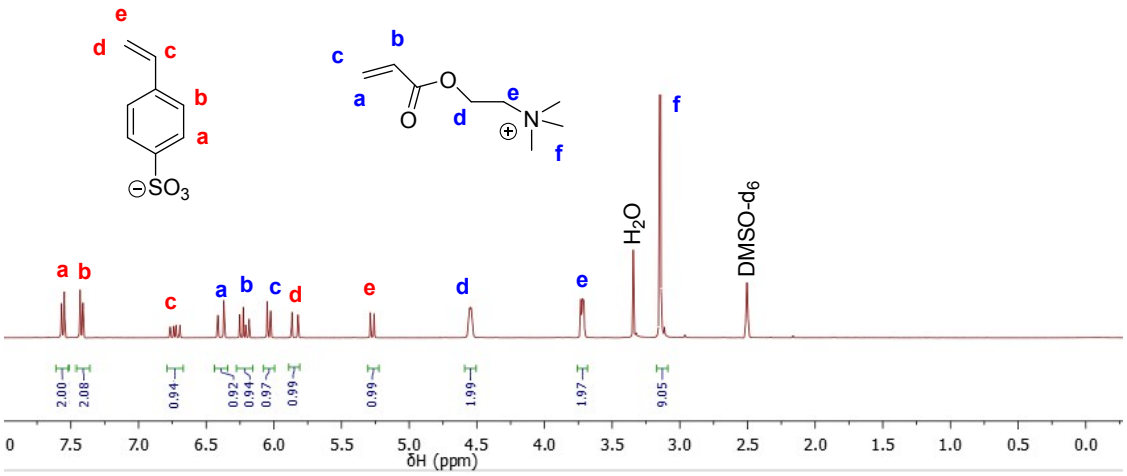

Fig. S2 ^{1}H NMR of the IPC salt **2**

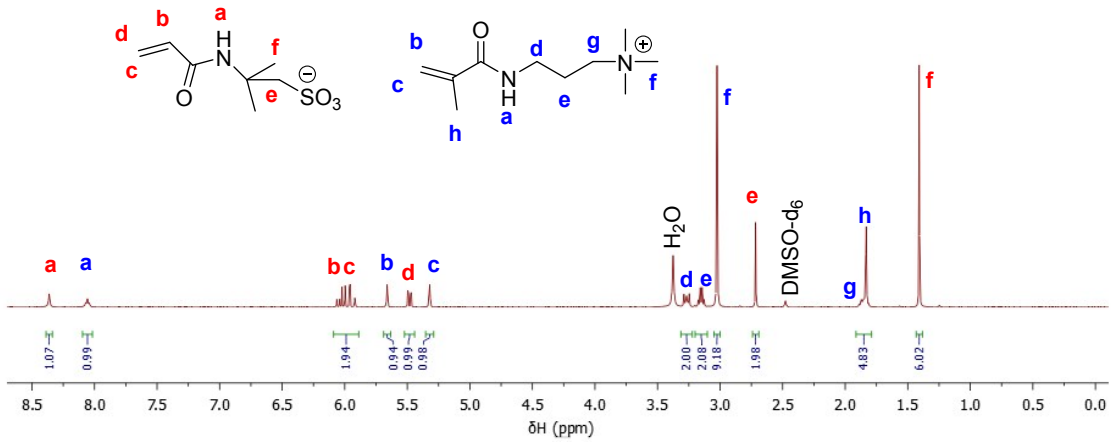

Fig. S3 ^{1}H NMR of the IPC salt **3**

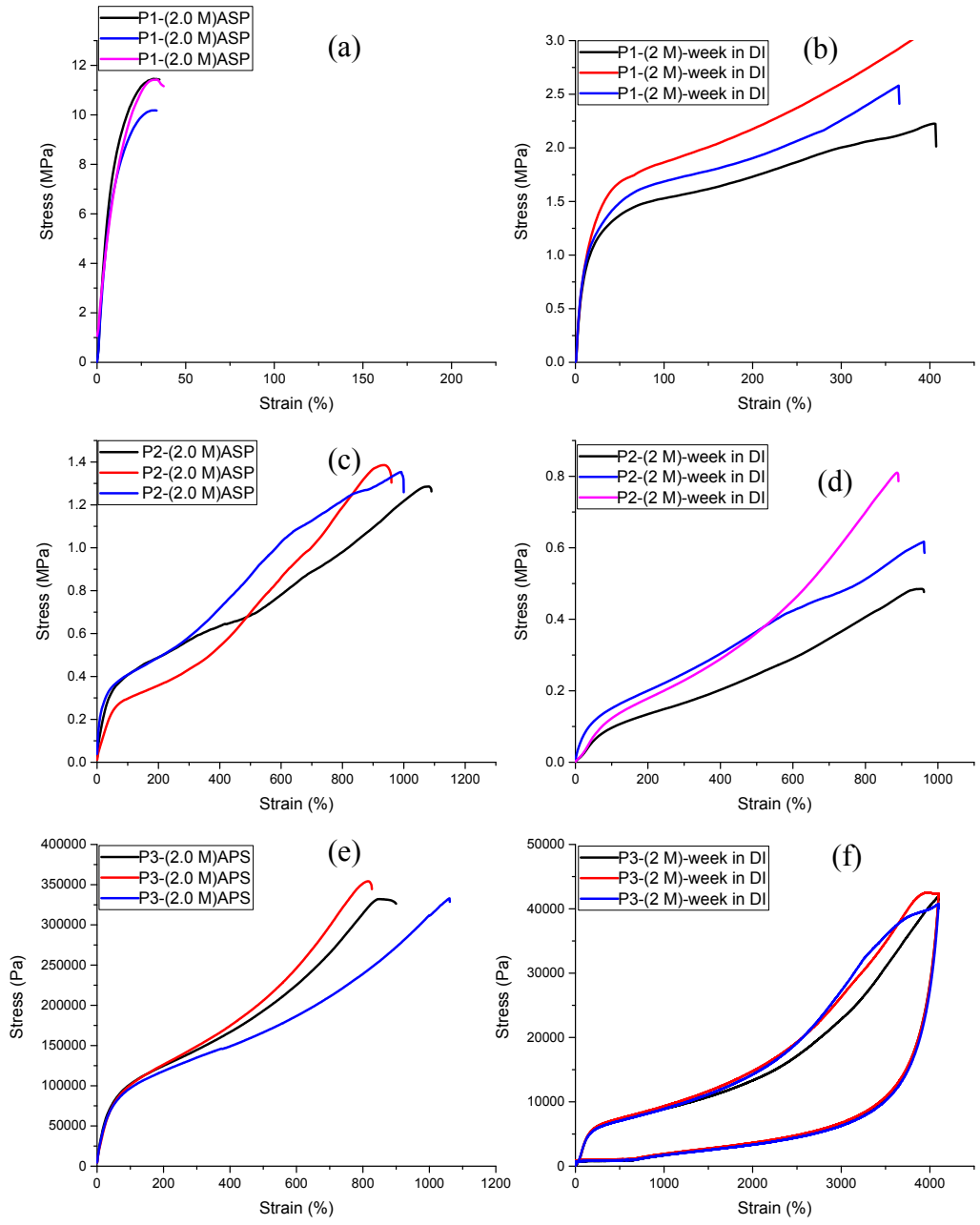

Fig. S4 Three replicates of tensile tests, (a), (c) and (e) illustrate the 3 replicates of ASP gels of **P1**, **P2** and **P3** and (b), (d) and (f) are illustrate the same gel after equilibrate for week in DI water.

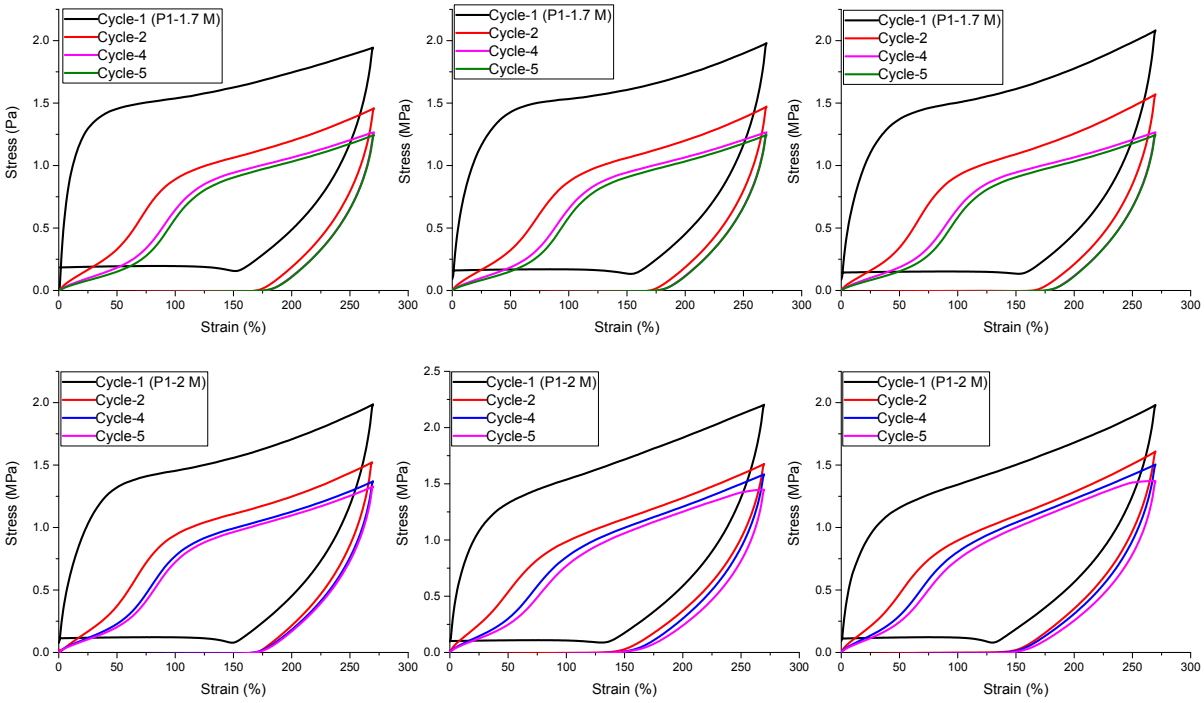

Fig. S5 Three replicates of cycling tests, gel **P1(1.7 M)** cycling at 27 mm (top), gel **P1(2 M)** cycling at 27 mm (bottom) for 5 cycles with 10 minute recovery time between cycles.

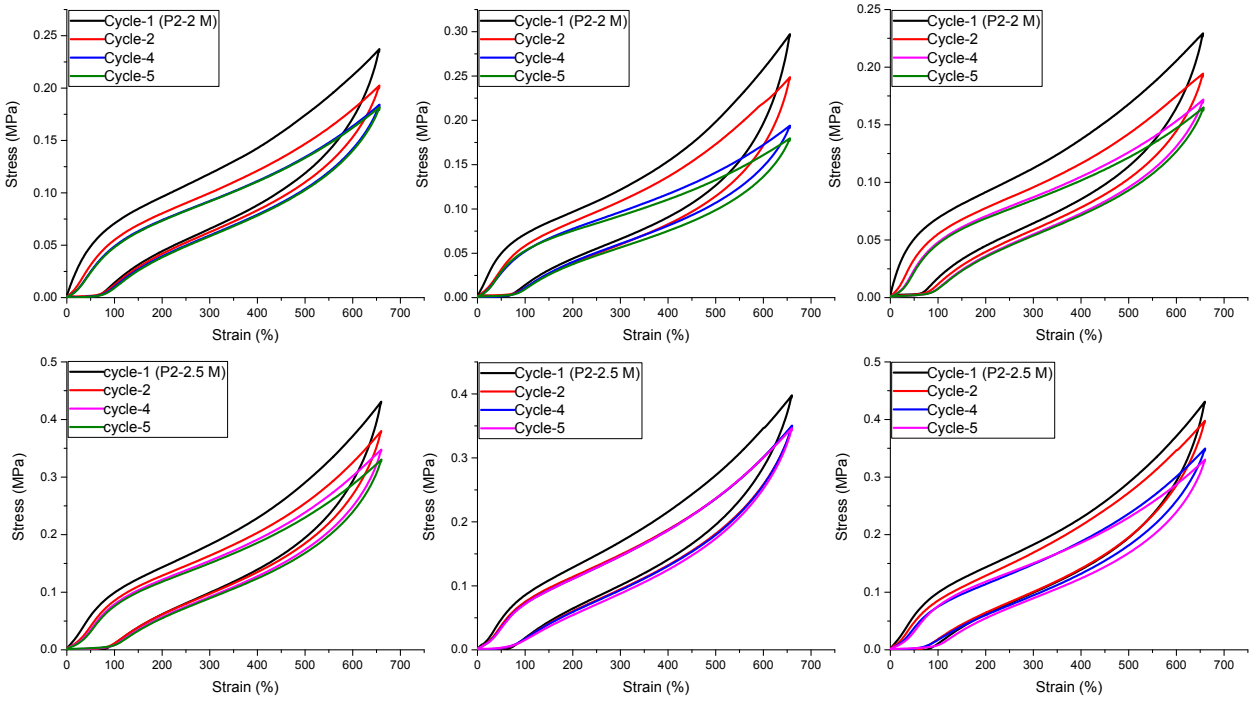

Fig. S6 Three replicates of cycling tests, gel **P2(2 M)** cycling at 66 mm (top), gel **P2(2.5 M)** cycling at 66 mm (bottom) for 5 cycles with 10 minute recovery time between cycles.

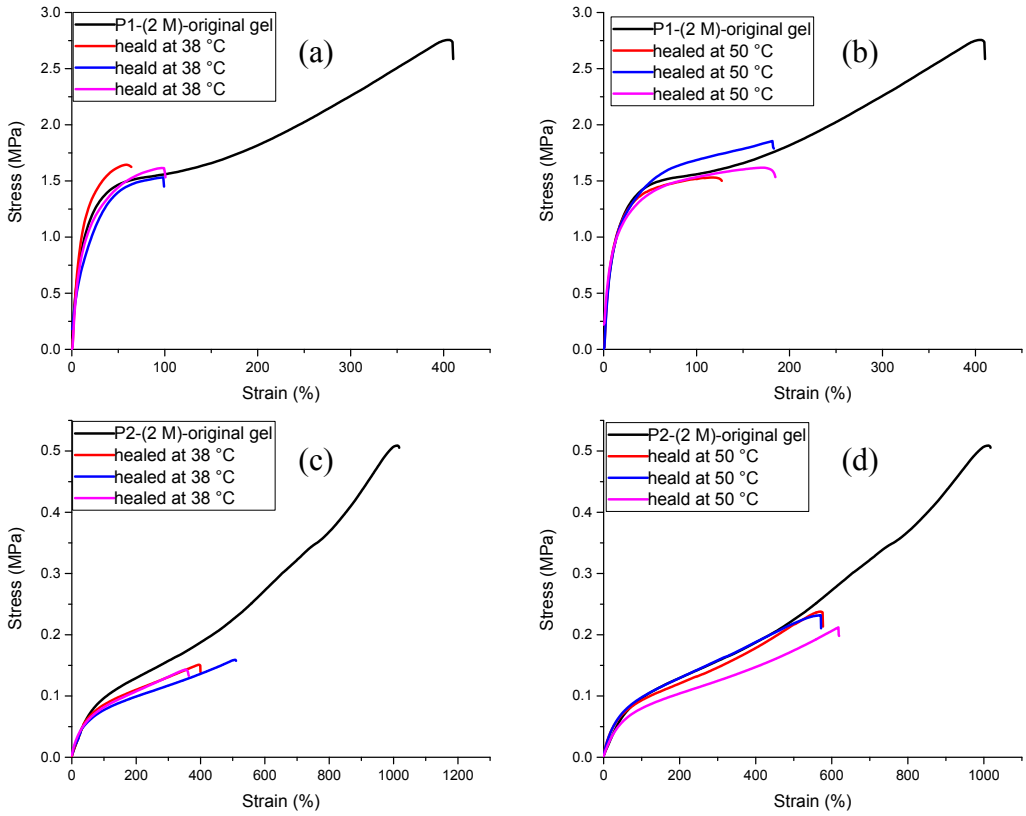

Fig. S7 Three replicates of healing tests at elevated temperatures in DI water. (a), (b) healing behaviour of gel **P1(2 M)** at 38 °C and 50 °C, (c), (d) healing of gel sample **P2(2 M)** at same temperature respectively.

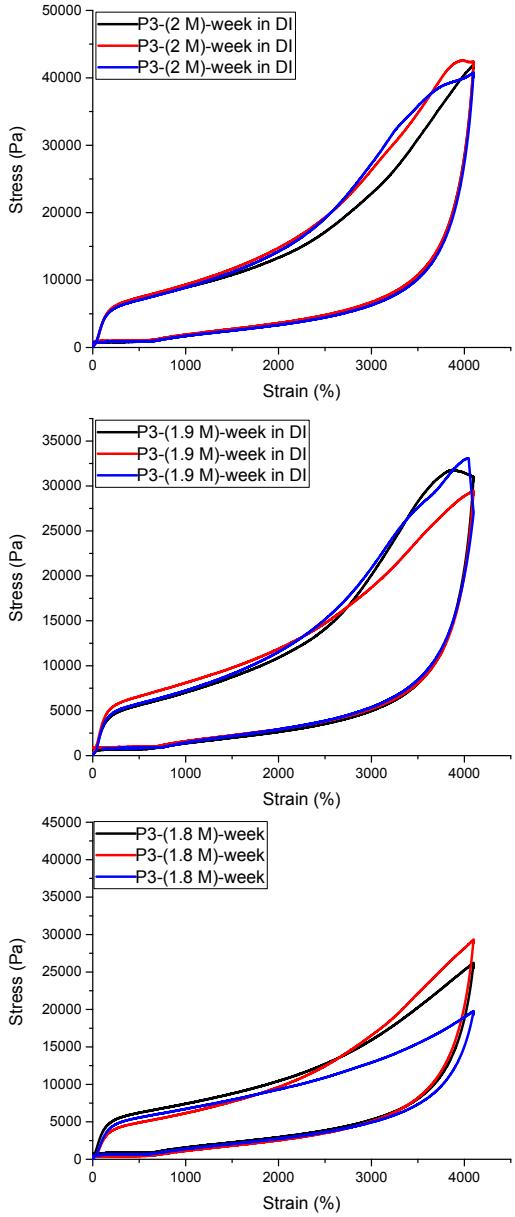

Fig. S8 Three replicates of tensile tests of gel **P3** at different monomer concentrations.

Observed mechanical properties in the three types of polyampholyte gels are summarized in the table 2 and 3.

Table. S1 The summary of mechanical properties of polyampholyte hydrogels type of **P1** and **P2**.*

Gel type	C (M)	σb (MPa)	εb (mm/mm)	E (MPa)	$A1^{st}$ /$A5^{th}$ (%)
P1-APS	2.0	11.02 ± 0.72	3.3 ± 0.06	9.63 ± 1.48	-
P1-wek	1.7	1.94 ± 0.23	35.30 ± 1.32	0.60 ± 0.025	-
P1-wek	2.0	2.53 ± 0.27	38.58 ± 2.0	1.04 ± 0.02	51.49 ± 2.72
P2-wek	2.0	1.34 ± 0.05	100.2 ± 7.3	0.054 ± 0.01	-
P2-wek	2.0	0.62 ± 0.15	93.41 ± 4.02	0.014 ± 0.005	75.11 ± 3.76
P2-wek	2.5	0.75 ± 0.085	82.24 ± 2.06	0.017 ± 0.0005	82.24 ± 1.24

* C, σb, εb, E, and $A1^{st}$/$A5^{th}$, represents the monomer concentration, fracture stress, fracture strain, Young's modulus and percentage recovery ratio calculated from hysteresis areas of the 1^{st} cycle vs 5^{th} cycle for the gel. The abbreviation (wek) mean gels are equilibrating in week time in DI water.

Table. S2 The summary of mechanical properties of polyampholyte hydrogels type of **P3**.*

Gel type	C (M)	σb (kPa)	εb (mm/mm)	E (kPa) (maximum)	$A1^{st}$ /$A5^{th}$ (%)
P3-wek	2.0	41.72 ± 0.83	>400	0.344 ± 0.011	at 280 mm 39.5 ± 1.53 at 100 mm 64.1 ± 1.85
P3-wek	1.9	29.19 ± 1.97	>400	0.298 ± 0.015	-
P3-wek	1.8	23.19 ± 0.27	>400	0.271 ± 0.04	-

* Abbreviations have same meaning as before.

Supplementary Movie SM 1

(Video is playing at 4× faster than the actual strain rate)