Supporting Information

Tough supramolecular hydrogels of poly(*N,N*-dimethylacrylamide)-grafted poly(methacrylic acid) with cooperative hydrogen bonds as physical crosslinks

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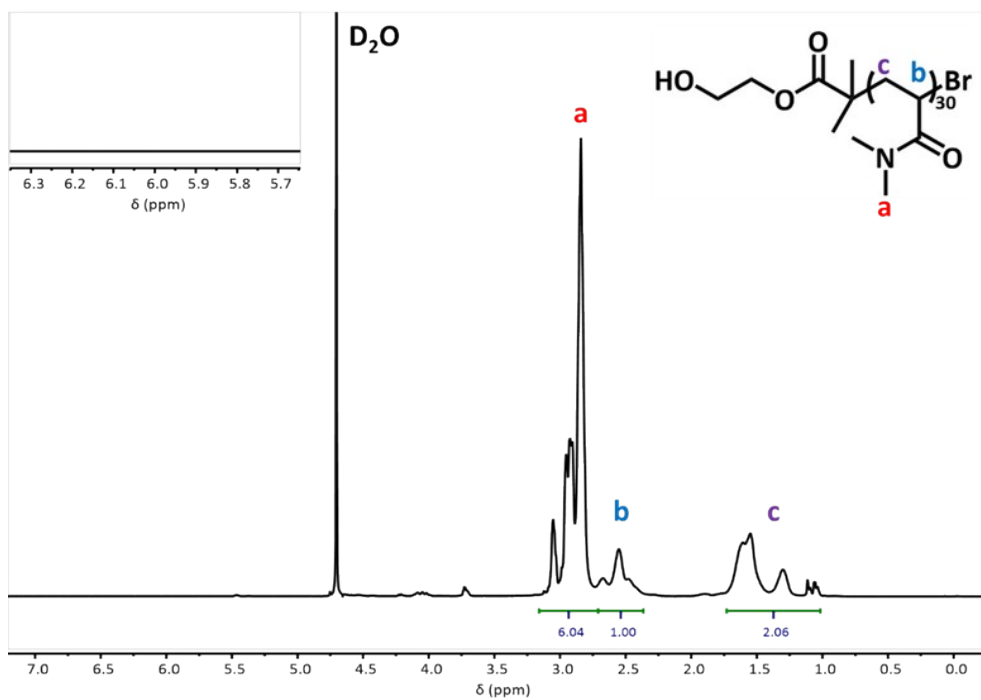*Corresponding authors. E-mail: cong.du@qdu.edu.cn, wuziliang@zju.edu.cn

Scheme S1 Synthesis of PDMAA oligomers and PDMAA macromonomers.

Fig. S1 Molecular weight distributions of PDMAA oligomers measured by size exclusion chromatography.

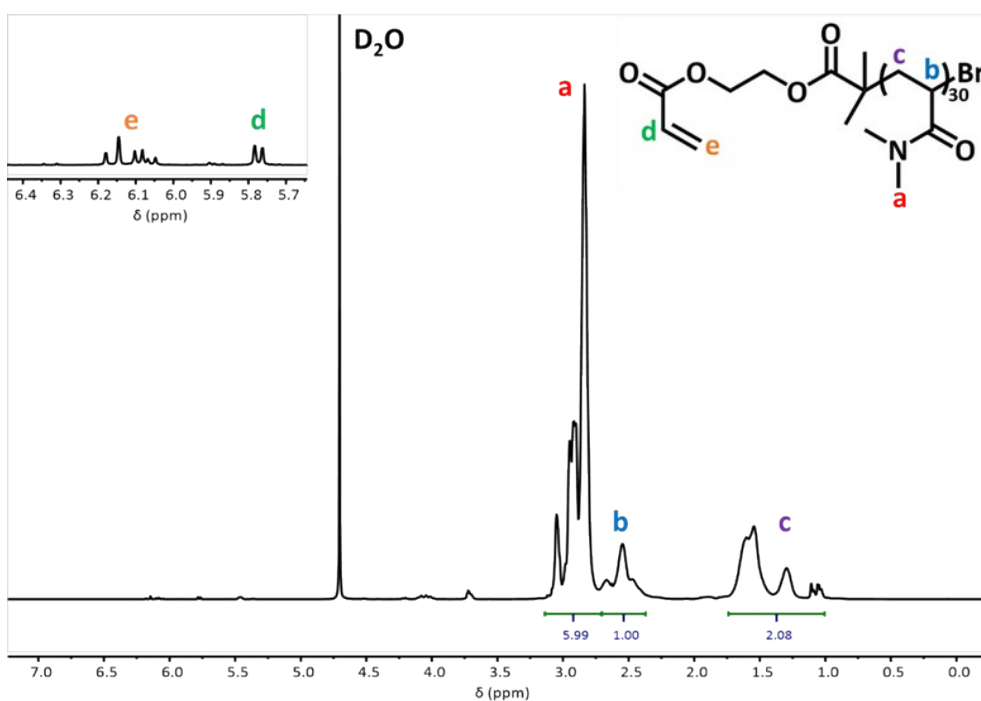**Fig. S2** ^{1}H-NMR spectrum of PDMAA oligomer by using D$_2$O (4.70 ppm) as the deuterated reagent.

Fig. S3 ^{1}H-NMR spectrum of PDMAA macromonomer by using D$_2$O (4.70 ppm) as the deuterated reagent.
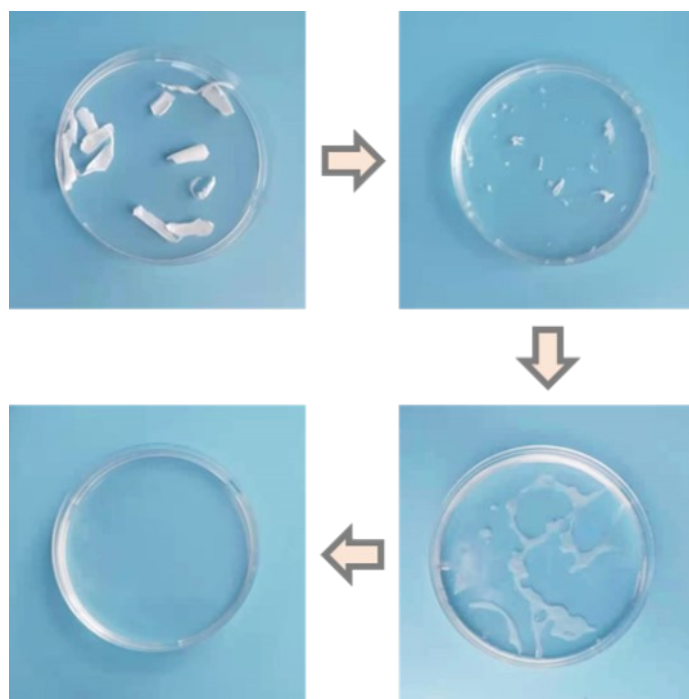
Fig. S4 The dissolution of GP-0.4 hydrogel in 1 M NaOH solution (pH = 14) at room temperature in 1 h.
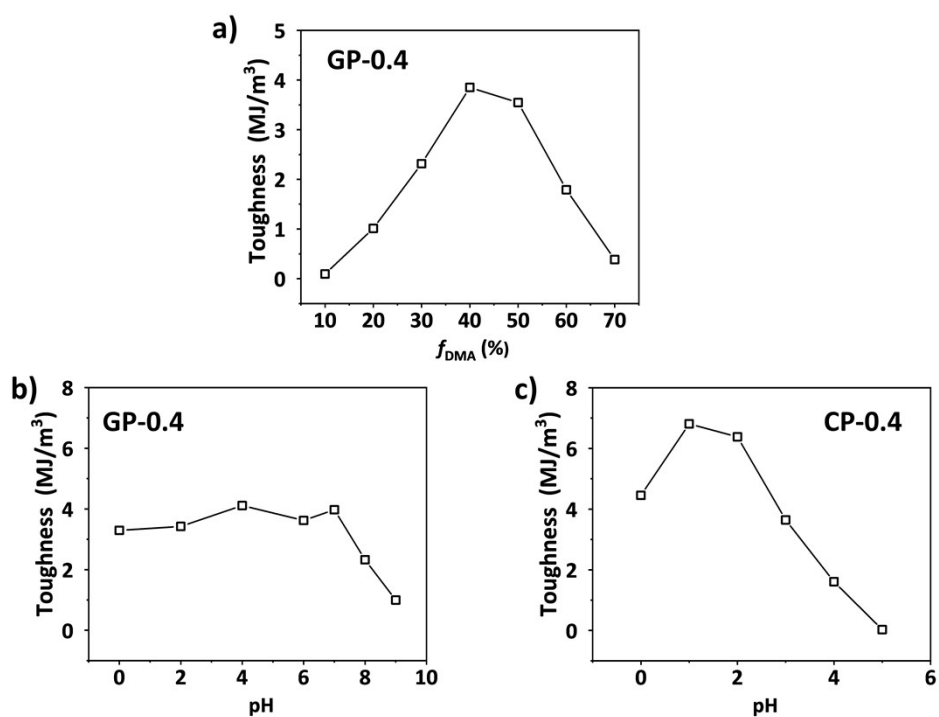

Fig. S5 (a) Toughness of the equilibrated GP hydrogels with different f. (b,c) Toughness of the GP-0.4 (b) and CP-0.4 (c) hydrogels equilibrated in aqueous conditions with different pH values at room temperature.
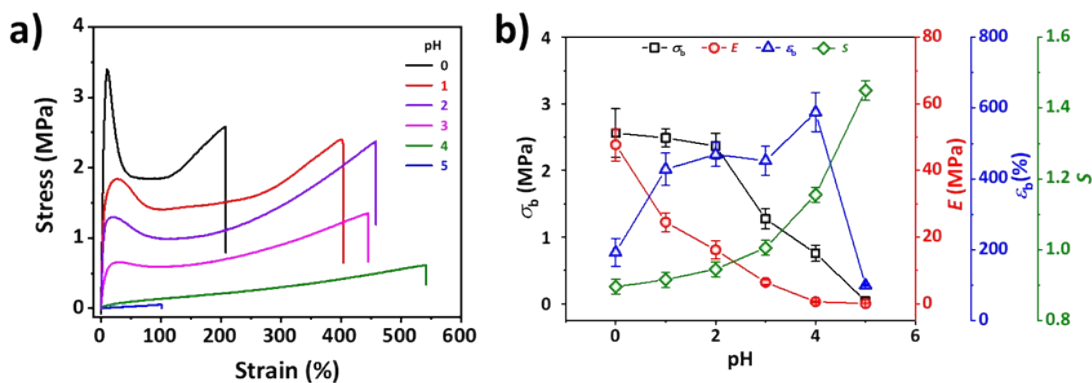
Fig. S6 Tensile stress-strain curves (a) and corresponding mechanical parameters (b) of the CP-0.4 hydrogels equilibrated in aqueous conditions with different pH values at room temperature. The swelling ratio in length, *S*, respected to the length of the gel at pH = 7 is also shown in (b).
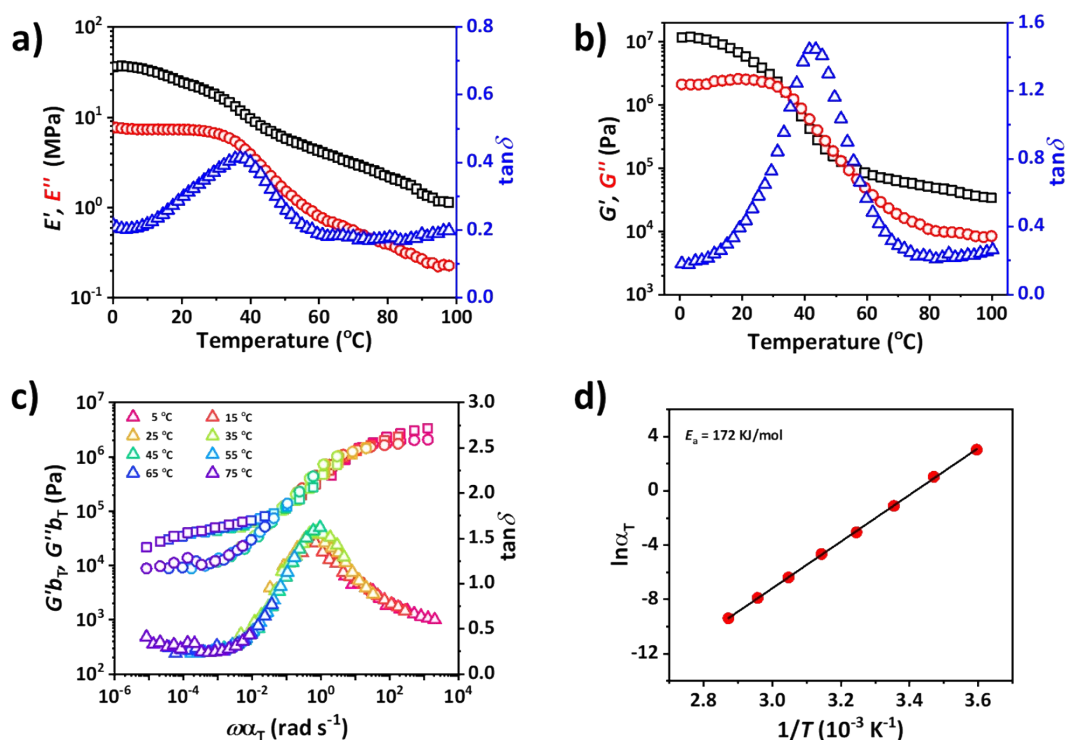

Fig. S7 (a) DMA spectra of the CP-0.4 gel with a heating rate of 1 °C/min. DMA measurement is performed in a single cantilever beam mode at a frequency of 1 Hz. (b) Temperature sweep of the GP-0.4 gel from 5 to 100 °C at a frequency of 5 Hz and strain amplitude of 0.2%. Heating rate: 5 °C/min. (c) Dynamic moduli and loss factor spectra of the GP-0.4 gel following TTS with the reference temperature of 20 °C. (d) Arrhenius plot of the horizontal shift factor α_T. The apparent activation energy E_a is calculated from the slop of the curve.

Table S1 The recipes of precursor solutions for GP hydrogels with different f.

Hydrogel	PDMAA-macromonomer [g]	MAAc [g]	ABVN [mg]	Solvent [g]
GP-0.1	0.6	5.4	85.38	13.91
GP-0.2	1.2	4.8	84.24	13.91
GP-0.3	1.8	4.2	83.10	13.92
GP-0.4	2.4	3.6	81.96	13.92
GP-0.5	3.0	3.0	80.84	13.92
GP-0.6	3.6	2.4	79.70	13.92
GP-0.7	4.2	1.8	78.56	13.92

Table S2 The recipes of the precursor solution for CP-0.4 hydrogels.

Hydrogel	DMAA [g]	MAAc [g]	APS [mg]	H_2O [g]
CP-0.4	2.4	3.6	75.31	13.92

Table S3 Elemental analysis results of the equilibrated GP and CP hydrogels. The top and bottom rows for each sample are the experimental value and theoretical value, respectively.

Hydrogel	N (wt%)	C (wt%)	H (wt%)
GP-0.1	1.24±0.08	55.77±0.22	7.50±0.01
	1.41	56.29	7.19
GP-0.2	2.31±0.04	56.32±0.37	7.51±0.09
	2.83	56.77	7.40
GP-0.3	3.99±0.08	56.92±0.35	7.71±0.04
	4.24	57.25	7.61
GP-0.4	5.35±0.07	57.68±0.41	8.00±0.03
	5.66	57.73	7.82
GP-0.5	6.66±0.07	58.54±0.50	8.10±0.19
	7.07	58.21	8.03
GP-0.6	7.96±0.05	58.07±0.01	8.16±0.25
	8.48	58.69	8.25
GP-0.7	9.03±0.09	59.86±0.10	8.07±0.35
	9.90	59.17	8.46
CP-0.4	5.91±0.09	57.61±0.02	7.99±0.08
	5.66	57.73	7.82