Electronic Supplementary Material (ESI) for Materials Advances.

Supporting information

Stimuli-responsive Hydrogel Consisting of Hydrazide-Functionalized Poly(oligo(ethylene glycol) methacrylate) and Dialdehyde Cellulose Nanocrystals

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Calculation of the theoretical molecular weight of POEGMA-H copolymer:

The theoretical molecular weight of POEGMA-H was estimated by Eq. S1:

([MEO2MA]+[OEGMA300]+[MAA])/[2f-BiB] × average molecular weight of monomers × conversion (S1)

where the total mass of monomers that are converted to polymer is divided by the mole of initiator (one initiator molecule is present in each chain) to calculate the average molecular weight of the chains.

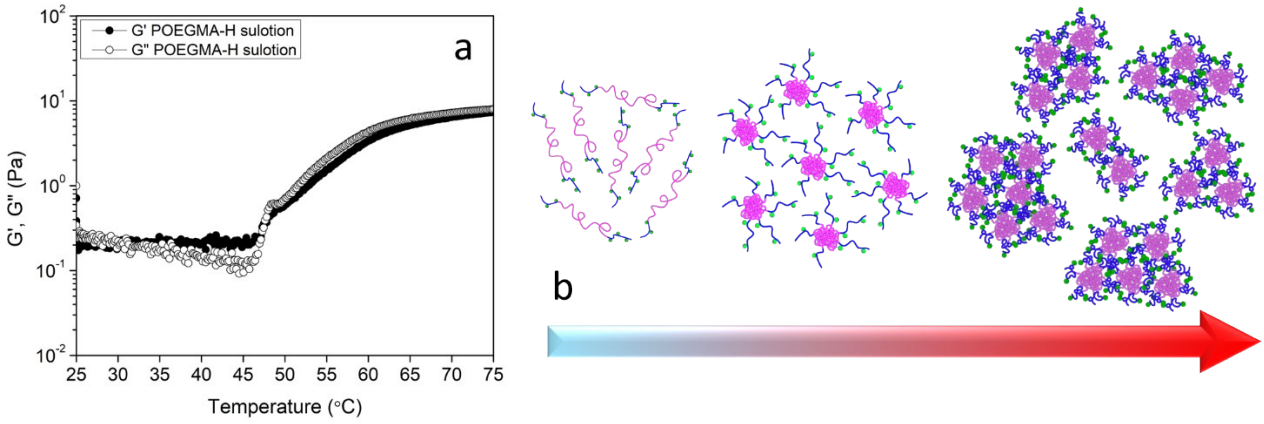

Figure S1. (a) Temperature sweep test of 20 wt % POEGMA-H aqueous solution at 1 Hz and 1 % strain. (b) Schematic representation of coil to globule transitions for middle and end blocks of POEGMA-H chains. Number of collapsed micelles presented in one cluster is more than what is shown here.

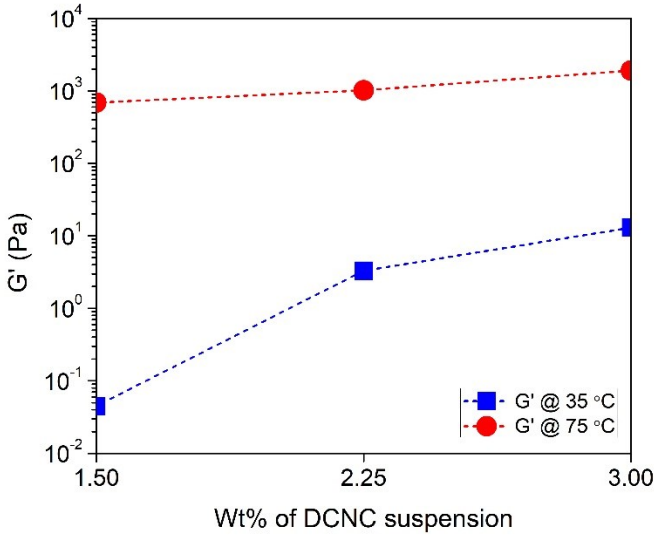

Figure S2. Effect of concentration of DACNC on the storage modulus of hydrogel before and after phase transitions.

Table S1. Estimates of β along with their associated temperature ranges for Gel1.5M, Gel2.25M, and Gel3M. Range of β is based on 95% confidence interval. ($\ln G_0 = constant + \ln T + 2\beta T$)

wt % of DCNC suspension	**Temperature range (°C)**	β	R^2
1.5	45-46	1.51 ± 0.221	0.988
	50-50	0.0799 ± 0.002	0.992
	62-65	0.294 ± 0.013	0.993
	68-75	0.006 ± 0.0003	0.984
2.25	26-30	0.005 ± 0.0001	0.998
	45-47	0.253 ± 0.018	0.989
	50-60	0.068 ± 0.001	0.992
	65-67	0.137 ± 0.007	0.994
	68-75	0.055 ± 0.001	0.995
3	26-30	0.007 ± 0.0003	0.989
	46-47	0.233 ± 0.003	0.999

	60-75	0.043 ± 0.0003	0.998

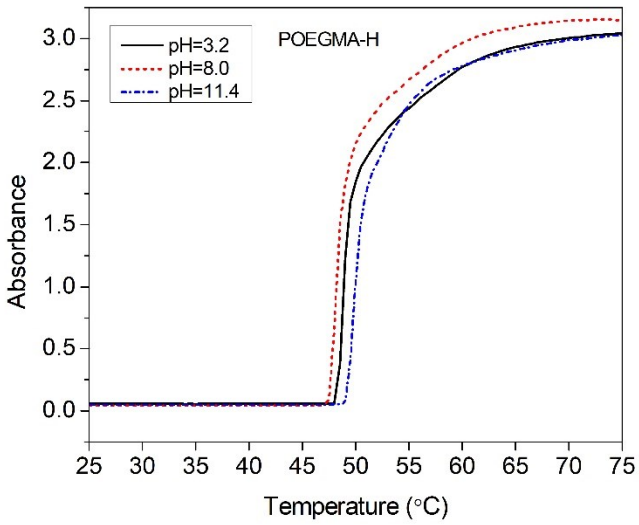

Figure S3. Effect of pH on phase transition of POEGMA-H aqueous solution (5 mg L^{-1}).

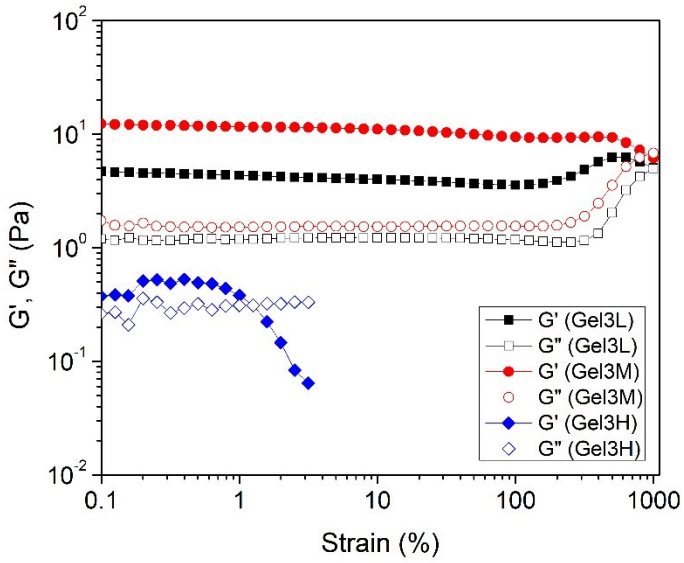

Figure S4. Linear viscoelastic region of Gel3L, Gel3M, and Gel3H at room temperature and 1Hz.

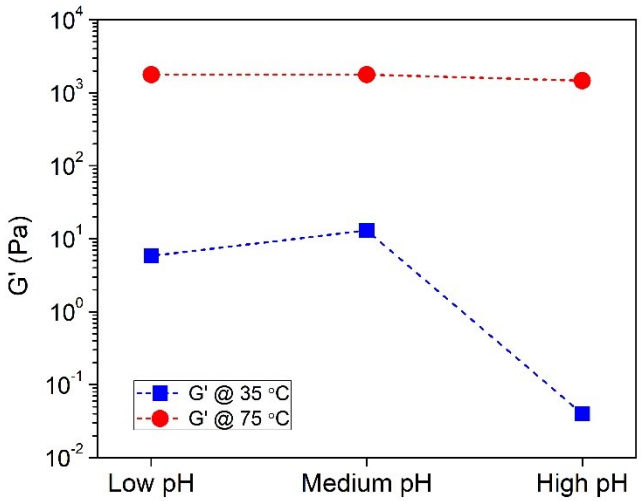

Figure S5. Effect of pH on the storage modulus of hydrogel before and after phase transitions.