

# SUPPORTING INFORMATION

## **Thermoresponsive composite hydrogels with aligned macroporous structure by ice-templated assembly**

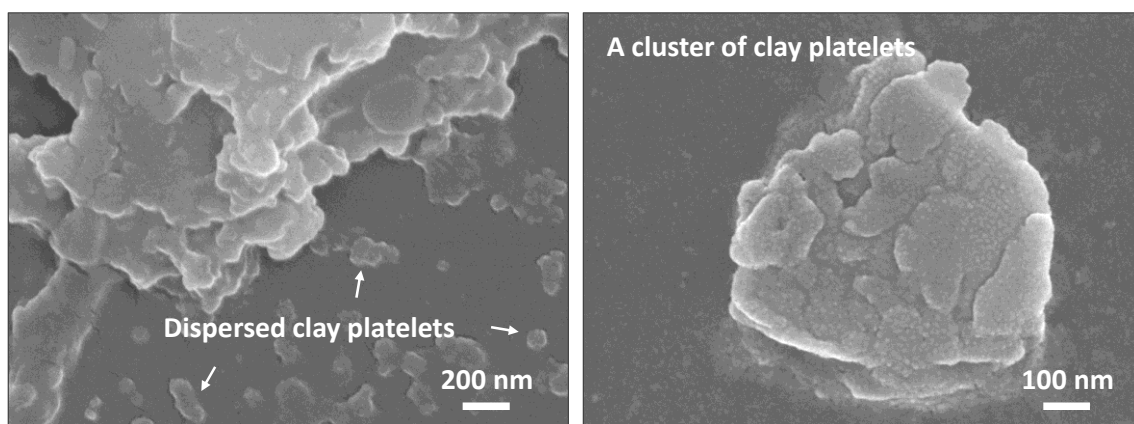
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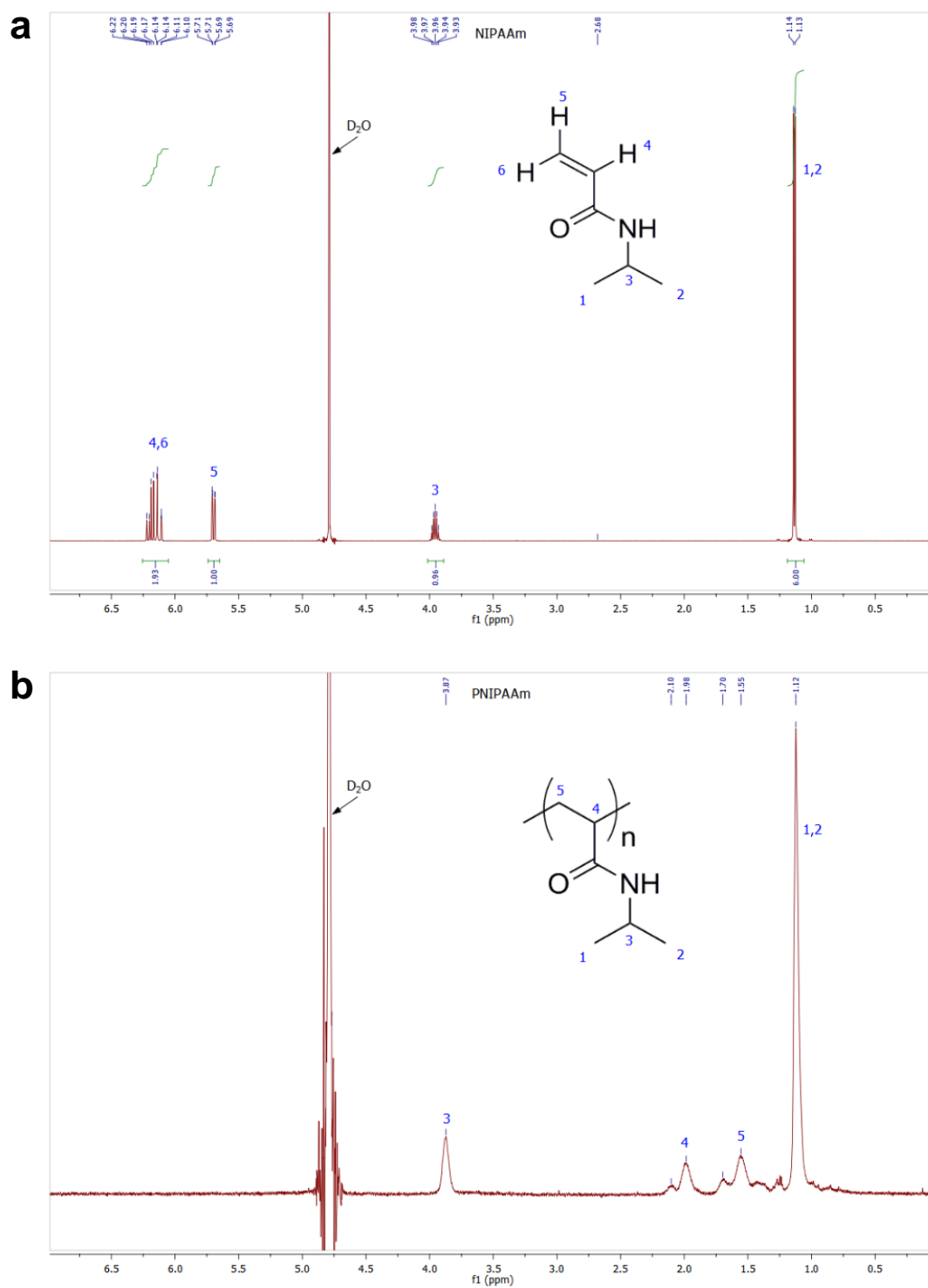
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**Table S1.** Composition of the solutions with the same amount of monomer (1 mol/L) and initiator (0.02 mol/L), but with different amounts of clay (5, 10, and 15  $\times 10^{-2}$  mol/L).

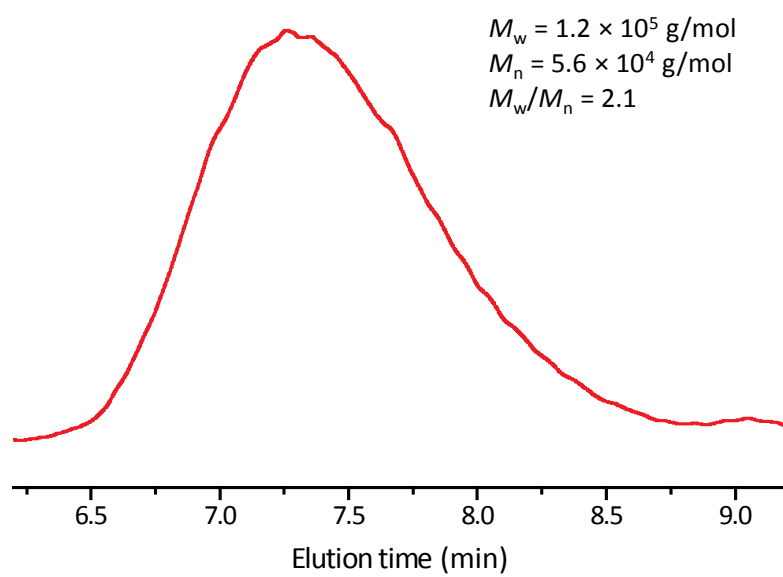
	NIPAAm (mol/L)	Clay (mol/L)	DEOP (mol/L)	Water (wt %)	NIPAAm (wt %)	Clay (wt %)	Clay/(Clay+polymer)
NC-5	1	0.05	0.02	86.55	9.79	3.30	0.25
NC-10	1	0.1	0.02	83.78	9.48	6.39	0.40
NC-15	1	0.15	0.02	81.19	9.19	9.28	0.50



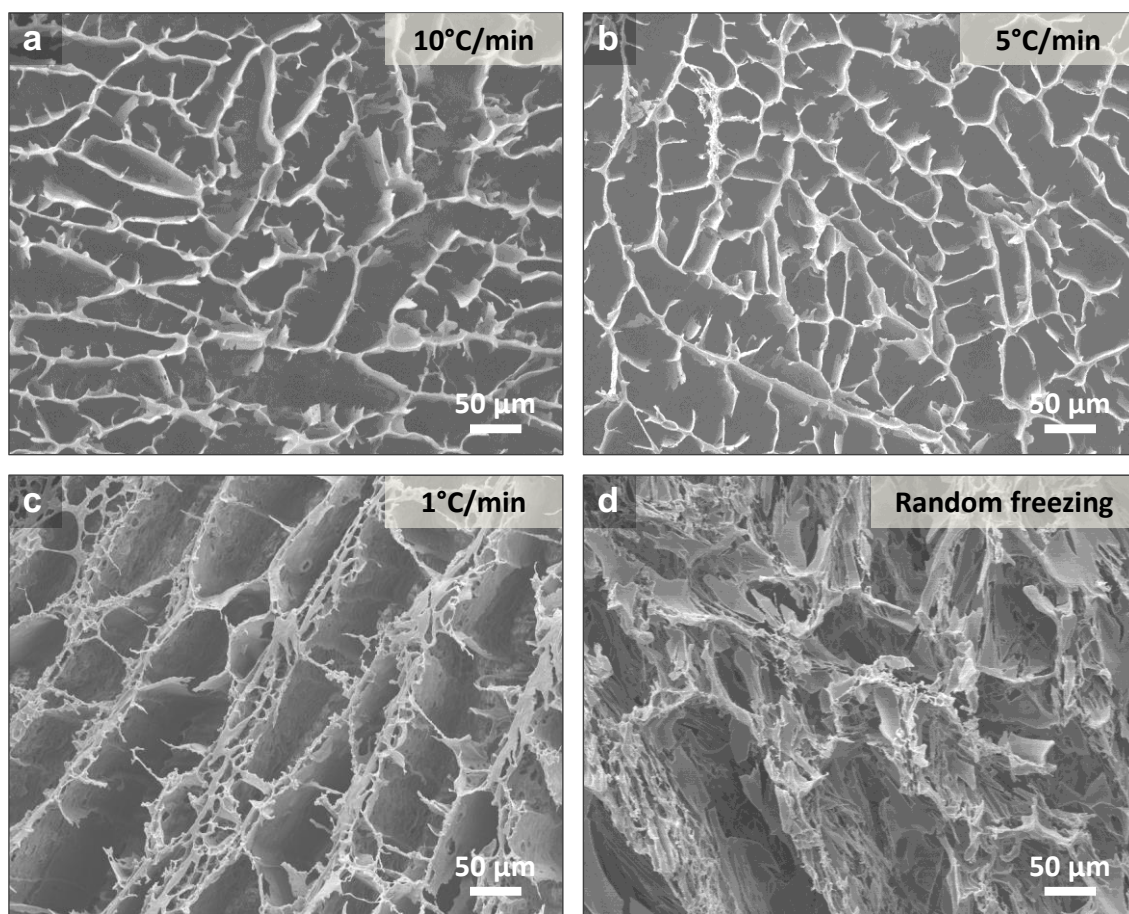
**Figure S1.** SEM images of clay platelets that have a diameter of  $\sim 100$  nm, and a thickness of  $\sim 1$  nm.



**Figure S2.**  $^1\text{H}$  NMR spectra of (a) NIPAAm and (b) PNIPAAm, which shows full conversion of monomer.



**Figure S3.** Gel permeation chromatography (GPC) measurement of PNIPAAm extracted through removing clay platelets.



**Figure S4.** SEM images of the samples (before polymerization) prepared by freeze-casting at a cooling rate of a) 10, b) 5, and c) 1 °C/min, or d) random freezing. Pore size is shown to increase with decreasing cooling rate.

**Table S2.** Mechanical properties (both parallel and perpendicular to the freezing direction) of NC-10 hydrogels fabricated by freeze-casting at different cooling rates and random freezing. Modulus was calculated from the period of stretch  $\lambda = 0 \sim 0.5$ , and fracture energy is calculated from the area under the stress-strain curve.

	Modulus (kPa)	Tensile strength (kPa)	Stretch (mm/mm)	Fracture energy (J/m <sup>2</sup> )
NC-10, Parallel, Random	45.6 ± 21.8	14.8 ± 4.1	6.9 ± 0.5	144.9 ± 157.9
NC-10, Parallel, 10°C/min	53.2 ± 16.2	114.5 ± 16.9	8.8 ± 2.1	4124.1 ± 2503.4
NC-10, Parallel, 5°C/min	41.2 ± 17.1	106.3 ± 19.8	11.2 ± 2.41	5092 ± 1732.4
NC-10, Parallel, 1°C/min	38.1 ± 7.7	79.4 ± 22.9	10.2 ± 1.7	4212.9 ± 1118.4
NC-10, Perpendicular, Random	42.6 ± 3.7	23.1 ± 3.9	9.2 ± 0.5	1094.4 ± 320.3
NC-10, Perpendicular, 10°C/min	41.1 ± 7.3	11.3 ± 0.3	5.9 ± 0.6	217.7 ± 20.4
NC-10, Perpendicular, 5°C/min	24 ± 0.4	18.2 ± 6.3	12.1 ± 0.2	450.9 ± 164.2
NC-10, Perpendicular, 1°C/min	21.9 ± 20.4	20.9 ± 2.8	17.5 ± 1.5	1553.7 ± 386.9