ESI Support Information

Gradient porous PNIPAM-based hydrogel actuators with rapid response and flexibly controllable deformation

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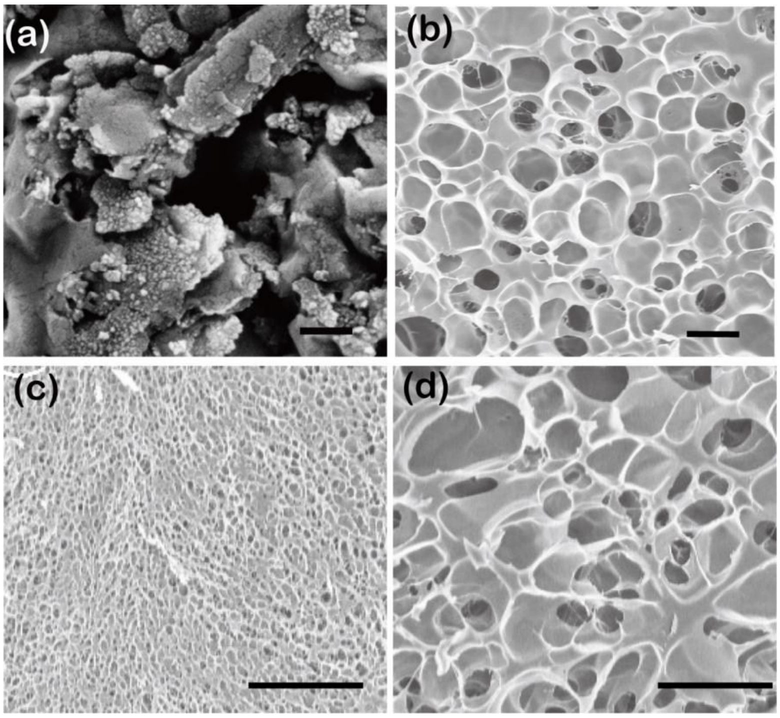

Figure S1. (a) SEM image of MMT particles. The scale bar was 200 nm. (b) Microscopic pore structures of pure PNIPAM hydrogel. The scale bar was 2 μm. (c) and (d) Microscopic pore structures of 0.2MN composite hydrogel in the bottom side and top side, respectively. The scale bars were 10 μm.

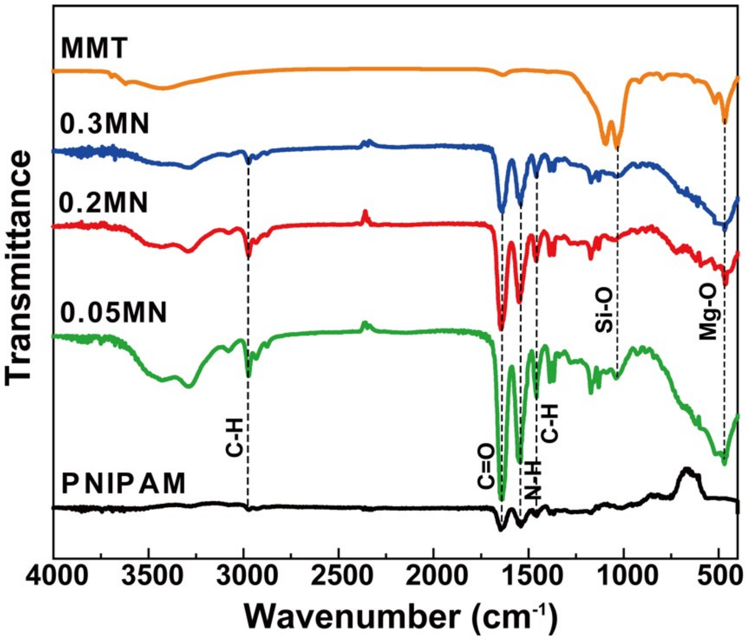

Figure S2. FT-IR spectra of MMT, 0.05MN, 0.2MN, 0.3MN and pure PNIPAM hydrogel.

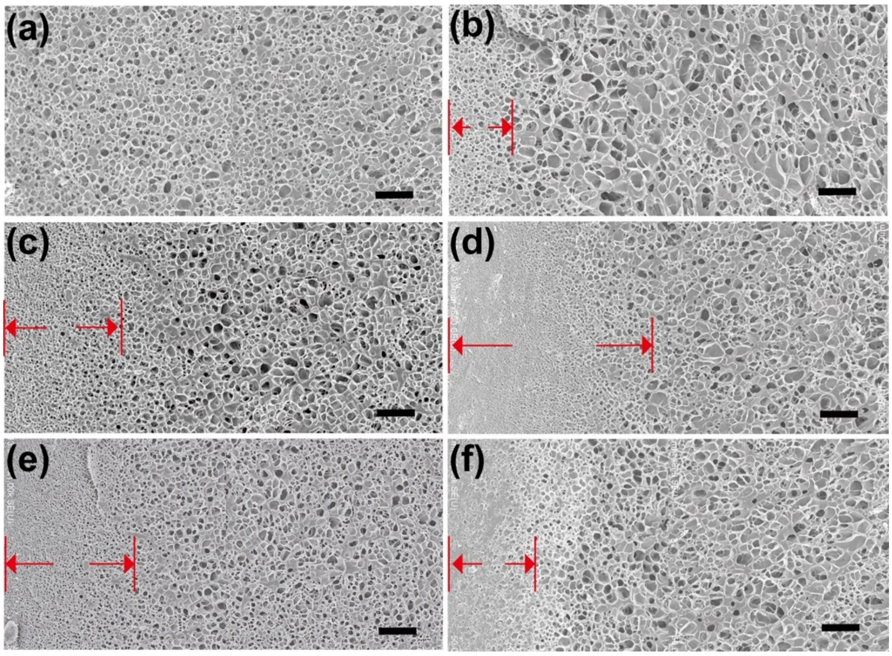

Figure S3. SEM images in the cross section of freeze-dried (a) pure PNIPAM, (b) 0.05MN, (c) 0.1MN, (d) 0.2MN, (e) 0.25MN and (f) 0.3MN composite hydrogels with large-ranged gradient structures along the direction of the gravity. The scale bars were 10 μm.

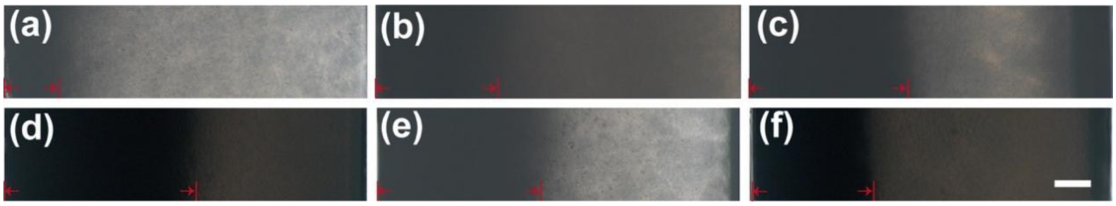

Figure S4. Optical microscope images for the cross section of (a) 0.05MN, (b) 0.1MN, (c) 0.15MN, (d) 0.2MN, (e) 0.25MN and (f) 0.3MN composite hydrogels. The scale bars were 100 μm.

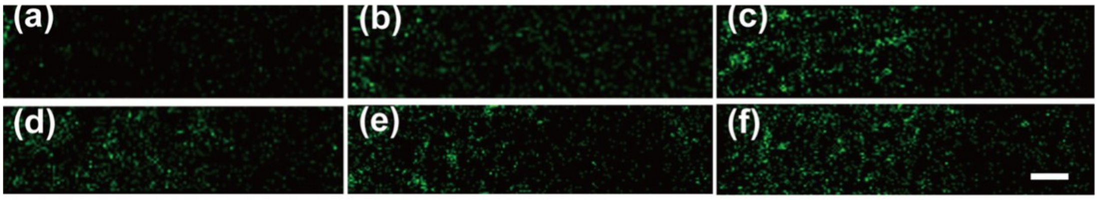

Figure S5. Si elemental mapping for the cross section of freeze-dried (a) 0.05MN, (b) 0.1MN, (c) 0.15MN, (d) 0.2MN, (e) 0.25MN and (f) 0.3MN composite hydrogels. The scale bars were 100 μm.

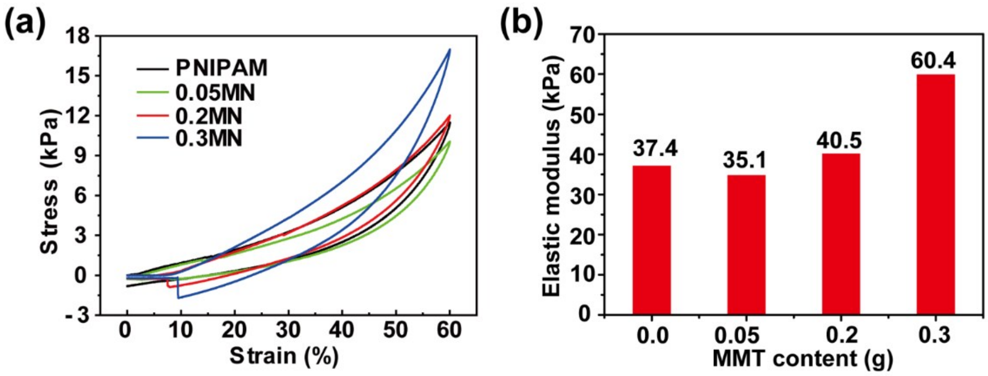

Figure S6. (a) Compress stress-strain curves of the hydrogels with different MMT contents and (b) the elastic modulus of different hydrogels.

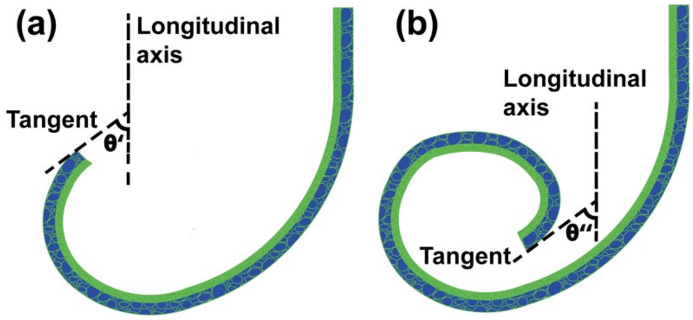

Figure S7. Schematic measurement for bending angle of the curled hydrogel.

When the hydrogel strip bends more than 180 ° (Figure S7a), the bending angle θ will be expressed as

$$\theta = \theta^{'} + 180°. \tag{S1}$$

If the hydrogel strip bends more than 360 ° (Figure S7b), the θ will be expressed as

$$\theta = \theta^{''} + 360°. \tag{S2}$$

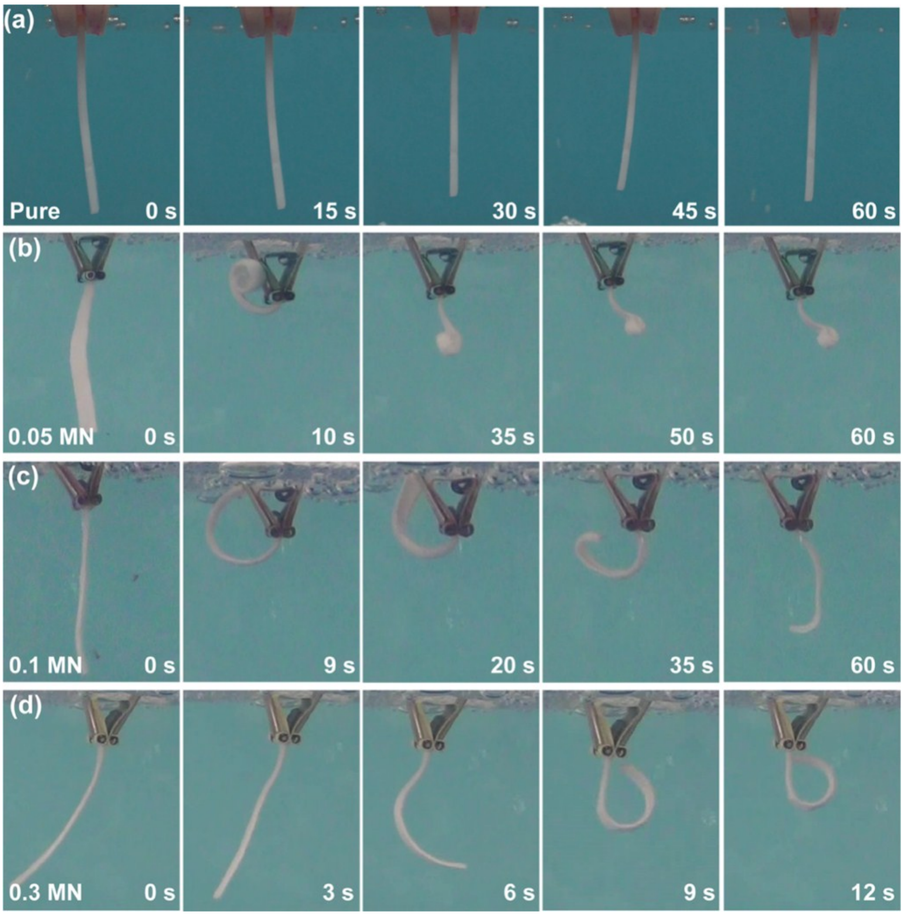

Figure S8. The bending behaviour of pure hydrogel, 0.05MN, 0.1MN and 0.3MN composite hydrogels in water at 50 °C, respectively. The thicknesses of all hydrogels were 1 mm.

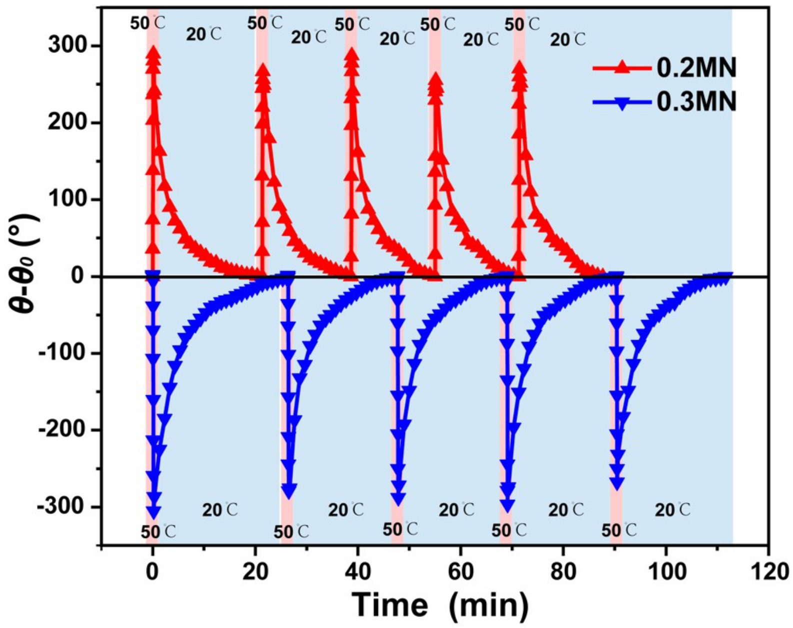

Figure S9. Reversible response and recovery of 0.2MN and 0.3MN composite hydrogels in water at 50 °C and 20 °C, respectively. (0.2MN with bidirectional bending characteristics only records the stage 1 bending process in water at 50 °C. θ_0 was the initial bending angle of the hydrogel strip immersed in water at 50 °C.

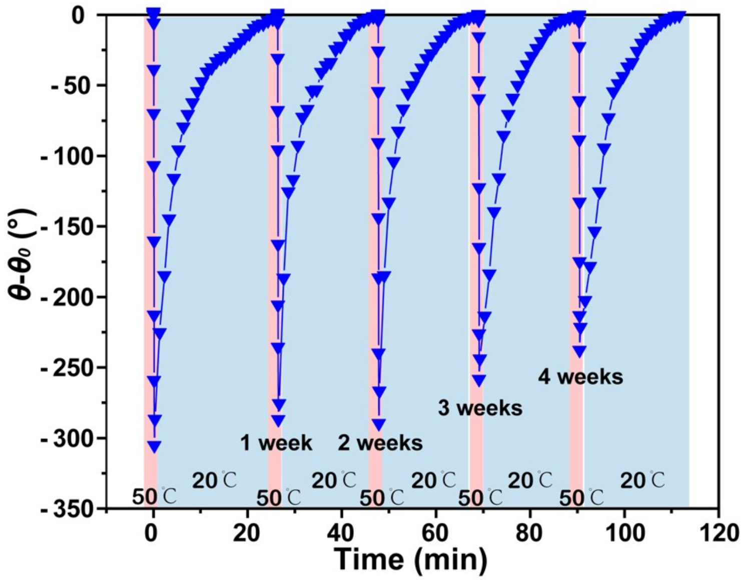

Figure S10. Long-term bending-recovery cycle test of 0.3MN composite hydrogel at 50 °C and 20 °C in water. θ_0 was the initial bending angle of the hydrogel strip immersed in water at 50 °C.

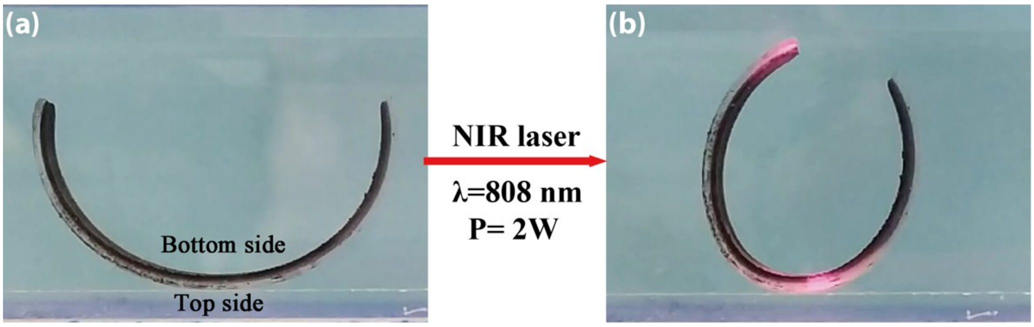

Figure S11. (a)-(b) Laser irradiation induced bending of 0.3MN composite hydrogel strip in water at 20 °C when CNTs were added into composite hydrogel.

Table S1. Parameter comparation of different hydrogel actuators in Figure 4c.

Hydrogel system	Anisotropic structure	Stimulating way	Response time (s)	Bending amplitude (º)	Bending velocity (º/s)	Ref.
PNIPAM	Gradient	Temperature	9	259	28.8	Our work
PNIPAM	Bilayer	Temperature	50	140	2.8	3
PNIPAM	Bilayer	IR irradiation	90	80	0.9	10
PNIPAM	Bilayer	Temperature	20	108	5.4	12
PNIPAM	Gradient	Temperature	16	221	13.8	13
PAA	Bilayer	pH	75	306	4.1	18
PNIPAM	Bilayer	Temperature	200	274	1.4	31
PNIPAM	Bilayer	Temperature	55	360	6.5	32
PNIPAM	Gradient	Temperature	21	105	5.0	35

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