

Supporting Information

Magnetic-field induced shape memory hydrogels for deformable actuators

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Synthesis of PMAAm hydrogels: Chemically cross-linked PMAAm hydrogel was prepared by free radical polymerization.^{S1} The precursor solution including 2 M MAAm, 0.5 mol % KPS, and 1 mol % MBAA (relative to the monomer) was added into the reaction cell, which was kept at 60 °C for 8 h to complete the reaction. The as-prepared hydrogel was swelled in a large amount of water for a week to remove the residuals and achieve the equilibrium state.

Synthesis of PMAAc hydrogels: Chemically cross-linked PMAAc hydrogel was prepared by free radical polymerization. The precursor solution including 2 M MAAc, 0.5 mol % KPS, and 1 mol % MBAA (relative to the monomer) was added into the reaction cell, which was kept at 60 °C for 8 h to complete the reaction. The as-prepared

hydrogel was swelled in a large amount of water for a week to remove the residuals and achieve the equilibrium state.

Synthesis of P(MAAm-co-MAAc) hydrogels: The P(MAAm-co-MAAc) hydrogel was synthesized by free radical copolymerization. Prescribed amounts of MAAm, MAAc, and KPS were dissolved in deionized water. After degassing for 30 min by bubbling of argon, the accelerator TEMED was added to the precursor solution, which was transferred into a reaction cell and kept at 60 °C for 8 h to complete the reaction. The as-prepared hydrogel was swelled in a large amount of water for a week to remove the residuals and achieve the equilibrium state.

Synthesis of PVA hydrogels: PVA hydrogel was synthesized through freezing-thawing cycle method.^{S2} Firstly, 10 wt% (relative to the mass of deionized water) PVA powder was dissolved into deionized water with gentle magnetic stirring for 2 h under 90 °C water bath to form PVA solution. Followed by vacuum treatment, the PVA solution was subsequently put into a container at -20 °C for 12 hours and thawed at room temperature for additional 12 hours. This freezing-thawing process was repeated 15 times to achieve the PVA hydrogel. All these as-prepared hydrogels were incubated in a large amount of deionized water for a week to remove the residuals and reach the equilibrium state.

Synthesis of P(MAAm-co-MAAc)/PVA hydrogels: The P(MAAm-co-MAAc)/PVA hydrogels were synthesized by free radical copolymerization. Prescribed amounts of MAAm, MAAc, PVA and KPS were dissolved in deionized water. After degassing for 30 min by bubbling of argon, the accelerator TEMED was added to the precursor solution, which was transferred into a reaction cell and kept at 60 °C for 8 h to complete the reaction. The as-prepared hydrogel was swelled in a large amount of water for a week to remove the residuals and achieve the equilibrium state.

References

S1 Y. J. Wang, X. N. Zhang, Y. Song, Y. Zhao, L. Chen, F. Su, L. Li, Z. L. Wu and Q. Zhang, *Chem. Mater.*, 2019, **31**, 1430–1440.

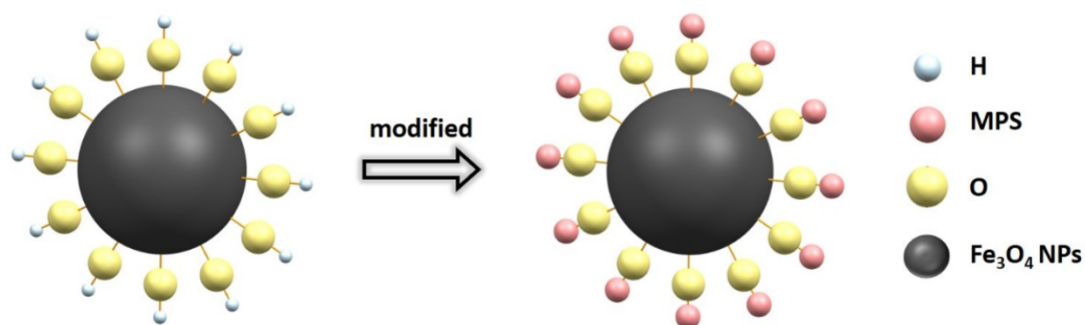


Figure S1 Schematic diagram of surface modification of Fe₃O₄ particles.

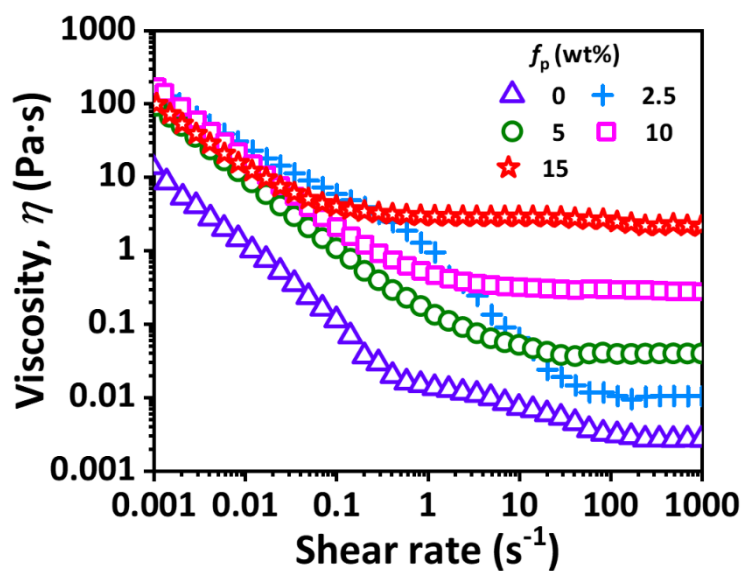


Figure S2 Viscosity of the ethanol solution of MMPFe-6-13- f_p -0 over a wide range of shear rate at room temperature.

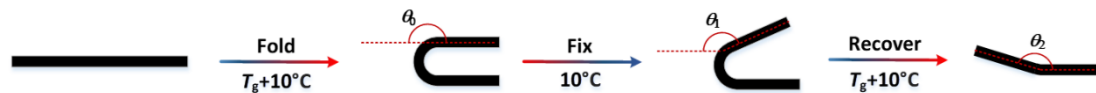


Figure S3 Schematic for the measurement of shape fixity ratio and shape recovery ratio of the gel by a folding mode.

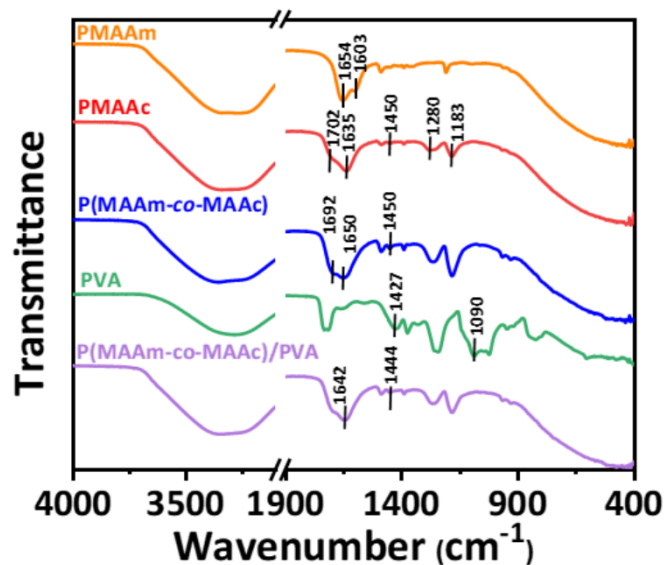


Figure S4 FTIR spectra of PMAAm, PMAAc, P(MAAm-co-MAAc), PVA and P(MAAm-co-MAAc)/PVA hydrogels.

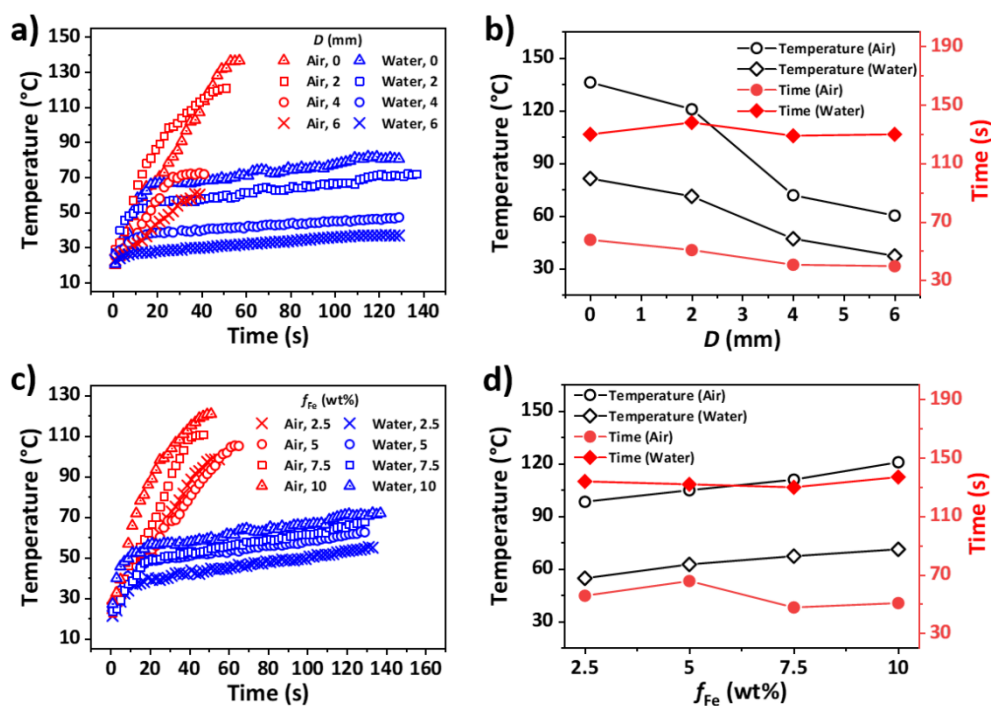


Figure S5 (a, b) The temperature increase of the magnetic hydrogel (MMPFe-6-13-10-10) under an AMF in water and air with different distance between hydrogel and the heating coil. (c, d) The temperature increase of the magnetic hydrogel (MMPFe-6-13-10-Fe) under an AMF in water and air with different Fe_3O_4 content (The magnetic hydrogel film is 0.2 mm away from the coil).

Table S1 Recipes of precursor solutions for synthesis of P(MAAm-co-MAAc)/PVA/Fe₃O₄ hydrogel films with different compositions.

Sample	Total volume (mL)	MAAm (g)	MAAc (g)	PVA (g)	KPS (g)	TMEDA (μL)	Water (g)	Fe ₃ O ₄ (g)
MMPFe-2-13-10-0	15	0.332	2.247	1.23	0.04	15	12.38	/
MMPFe-3-13-10-0	15	0.498	3.371	1.11	0.06	15	11.07	/
MMPFe-4-13-10-0	15	0.664	4.494	0.98	0.08	15	9.762	/
MMPFe-5-13-10-0	15	0.831	5.618	0.85	0.1	15	8.451	/
MMPFe-6-13-10-0	15	0.996	6.741	0.71	0.12	15	7.143	/
MMPFe-6-11-10-0	15	0.842	6.896	0.71	0.12	15	7.142	/
MMPM-6-12-10-0	15	0.919	6.812	0.73	0.12	15	7.269	/
MMPM-6-14-10-0	15	1.072	6.660	0.73	0.12	15	7.270	/
MMPM-6-15-10-0	15	1.149	6.586	0.73	0.12	15	7.271	/
MMPM-6-13-0-0	15	0.996	6.741	/	0.12	15	7.143	/
MMPM-6-13-2.5-0	15	0.996	6.741	0.18	0.12	15	7.143	/
MMPM-6-13-5-0	15	0.996	6.741	0.36	0.12	15	7.143	/
MMPM-6-13-10-0	15	0.996	6.741	0.71	0.12	15	7.143	/
MMPM-6-13-15-0	15	0.996	6.741	1.08	0.12	15	7.143	/
MMPM-6-13-10-2.5	15	0.996	6.741	0.71	0.12	15	7.143	0.375
MMPM-6-13-10-5	15	0.996	6.741	0.71	0.12	15	7.143	0.75
MMPM-6-13-10-7.5	15	0.996	6.741	0.71	0.12	15	7.143	1.125
MMPM-6-13-10-10	15	0.996	6.741	0.71	0.12	15	7.143	1.5