

Supporting information for
**Multiple shape memory polymers for self-
deployable device**

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1. Experimental Section

1.1 Raw materials

2-Methoxyethyl acrylate (MEA) and N-Methylol acrylamide (HAM) were purchased from Alfa Aesar, and used without further purification. Potassium persulfate (KPS) and N,N,N',N'-tetramethyldiamine (TEMED, Sinopharm Chemical Reagent, China) were used as received without further purification. The water used in all experiments was purified by a Millipore Direct-Q purification system and bubbled with N₂ for 1 h before use.

1.2 Preparation of samples

The appropriate amount of MEA and HAM were added in ultrapure water with N₂ gas bubbling for 1 h to remove any soluble O₂. After vigorous stirring for another 15 min, the solution was cooled to 0°C before catalyst (TEMED, 8 μL) and initiator (0.01 g in 0.5 ml water) were added. Finally, the final mixture was transferred to a tailored glass tube and polymerized at room temperature for 8h, subsequent at 60°C for 12 h to obtain the wet samples. The obtained samples were washed thoroughly with distilled water to remove the unreacted monomer and other impurities. The resulting samples were subsequently dried at room temperature for 24 h followed by drying for 24 h at 80°C under vacuum in order to obtain the samples.

The multiple shape memory polymers were synthesized in three steps. First, the mixture of MH30 was transferred to a glass tube, and let stand 20 min for partially polymerization. Then, the mixture of MH50 was added on top of the first region and let stand 20 min before the addition of the mixture of MH70. Finally, the obtained

mixture was polymerized at room temperature for 8h, subsequent at 60°C for 12 h. After dried, multiple shape memory polymers could be obtained.

The self-deployable device was synthesized as follows: first, the mixture was transferred to customized Teflon models (the two Teflon plates were separated by a double-sided, 2 mm thick adhesive tape spacer), then polymerized at room temperature for 8h, subsequent at 60°C for 12 h to obtain the wet film samples. The obtained samples were washed thoroughly with distilled water to remove the unreacted monomer and other impurities. After that, the samples were posited together in a certain order (MH30 on the top, MH50 in middle and MH70 at the bottom), and separated with each other by thin plastic films with a hole (10mm diameter) in the center, so that the wet samples could contact and bond together during drying. The assembled wet device was subsequently dried at room temperature for 24 h followed by drying for 24 h at 80°C under vacuum. After dried, self-deployable device could be obtained.

1.3 Measurements and characterizations:

FT-IR spectra were performed with a Nicolet 6700 instrument (Thermal Scientific, USA) by the KBr method in the range 500-4000 cm^{-1} . The mechanical properties of polymers were measured on an ASG-J electronic universal testing machine (Shimadzu Co., Japan) at room temperature. Clubbed samples with size of Φ 3 mm \times 40 mm (gauge length, 20 mm) were used for tensile tests with a crosshead speed of 20 mm/min. Plate samples with size of 40 mm \times 4 mm \times 0.5 mm was used for dynamic mechanical analysis with a TA Q800 Dynamic Mechanical Analysis (DMA)

instrument (TA Instrument Inc.) in the tensile mode. The dynamic storage and loss moduli were determined at a frequency of 10 Hz and a heating rate of 3°C/min as a function of temperature from -50°C to 100°C. DSC measurements were performed on a Seiko instrument (DSC 6200) in the temperature range -10~240 °C with a heating rate of 10 °C/min.

1.4 Shape memory performance:

Shape-memory property measurements were conducted according to the tensile tests and bending tests. In the tensile test, the samples of 40 mm length and 3 mm diameter were stretched to ε_m , 200% elongation at T_{high} (25°C for MH30, 55°C for MH50, and 80°C for MH70) with a 20 mm/min stretching rate (The gauge length is 20 mm, for observation, the sample was marked). Then, the clamps remained still, and the samples were cooled down to T_{low} (-16°C for MH30, 25°C for MH50, and 55°C for MH70). After 10 min, the samples were taken down from clamps, and the strain (ε_f) was measured. The shape recovery process was conducted at T_{high} for 10 min. The residual strain was defined as ε_r . The temperature of whole process was controlled using IR light and liquid nitrogen and measured using thermometer. The strain (ε), the shape fixing ratio (R_f) and the shape recovery ratio (R_r) were calculated by equations (1), (2) and (3) below:

$$\varepsilon = \frac{l - l_0}{l_0} \times 100\% \quad (1)$$

$$R_f = \frac{\varepsilon_f}{\varepsilon_m} \times 100\% \quad (2)$$

$$R_r = \frac{\varepsilon_m - \varepsilon_r}{\varepsilon_m} \times 100\% \quad (3)$$

where l and l_0 represent actual length and virgin length of the sample, respectively.

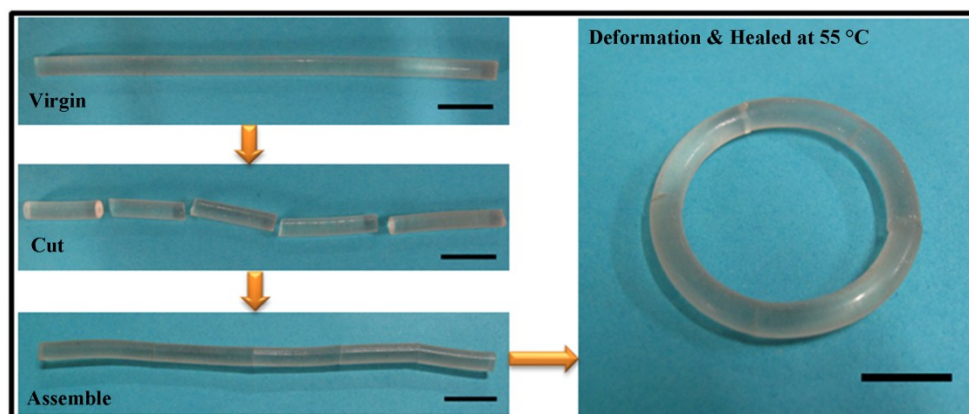


Figure S1. The real picture of MH50 sample assembled using five individual blocks.

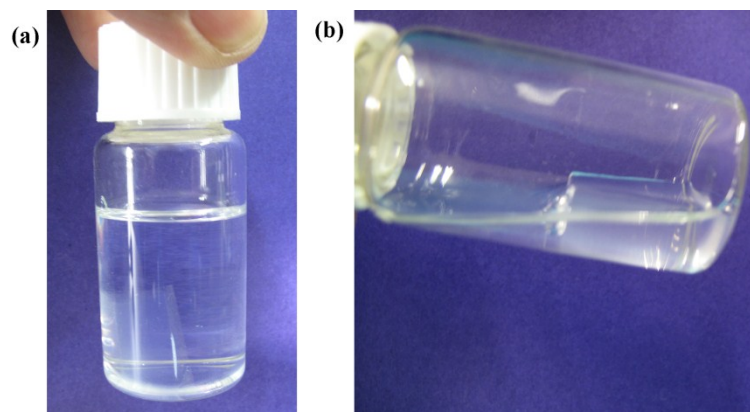


Figure S2. The sample of MH50 immersed into organic solvents after two months:

(a) THF; (b) Acetone.

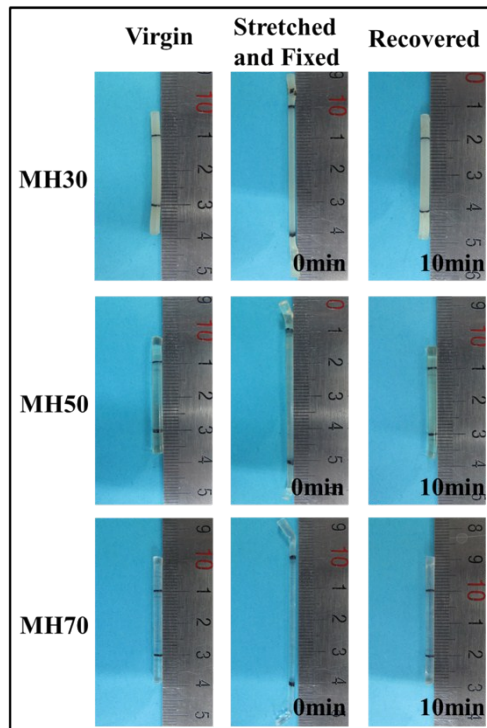


Figure S3. Shape memory effect of MH samples: MH30 was stretched and recovered at 25°C, fixed at -16°C; MH50 was stretched and recovered at 55°C, fixed at 25°C; MH70 was stretched and recovered at 80°C, fixed at 55°C.

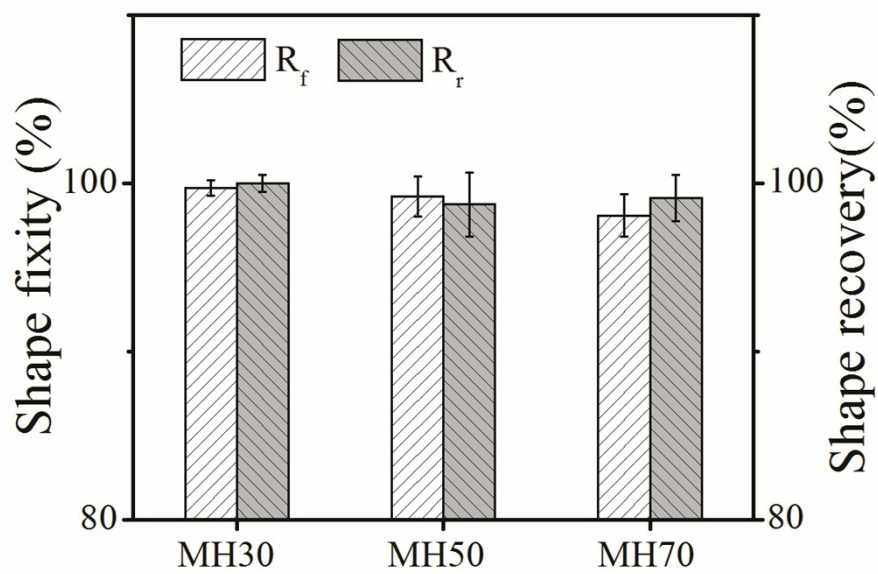


Figure S4. The shape fixity and recovery efficiencies of MH polymers.