Supporting information

Mimosa Inspired Bilayer Hydrogel Actuator Functioning in Multi-Environment

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Experimental Procedures

Materials: Acrylamide, *n*-hexane, acetone, ethanol, liquid paraffin, polyethylene glycol (M_W= 6000) were purchased from Sinopharm Chemical Reagent Co. Ltd. Acrylic acid, *N*-isopropyl acrylamide, *N,N′*-methylenebisacrylamide (BIS) (99%), rhodamine B (red dye), brilliant blue (blue dye) were purchased from Aladdin Reagent Co. Ltd. 2-Hydroxy-2-methylpropiophenone was obtained from the Sigma-Aldrich Company. *N*-isopropyl acrylamide was purified by recrystallization from mixing solvent of *n*-hexane and acetone prior use. Other reagents were used as received.

Fabrication of the bilayer hydrogel: The fabrication process is illustrated in Fig. S1. An AAm/AAc aqueous solution was prepared by mixing 0.5 g acrylamide, 1 g PEG and 500 μL acrylic acid in 8 mL deionized water, then 1 mL 0.01g/mL *N,N′*-methylenebisacrylamide aqueous solution and 4 μL 2-hydroxy-2-methylpropiophenone were added. After degased by N_2, the solution was sealed and stored in dark at 4°C.

The NIPAM aqueous solution was prepared by mixing 1 g *N*-isopropyl acrylamide, 1 g PEG in 8 mL deionized water, then 1 mL 0.01 g/mL *N,N′*-methylenebisacrylamide

aqueous solution and 4 μL 2-hydroxy-2-methylpropiophenone were added. After degased by N_2, the solution was sealed and stored in dark at 4°C.

In a typical polymerization process, AAm/AAc aqueous solution was injected into a silica rubber spacer attached to a glass slide. After covered by another slide of glass, the solution was irradiated under UV light (365 nm) for 3 min at 50°C in a water bath. Then the covering glass slide was lifted, a second silica rubber mold was put right above the first mold, NIPAM aqueous solution was injected into the mold and covered by the glass slide again. The mixture was then irradiated under UV light (365 nm) for 5 min in ice-water bath. After polymerization, the obtained bilayer hydrogel was washed by a large amount of water thoroughly. The size of glass slides used are 60 mm *60 mm* 3 mm and that of the silica rubber slices are 60 mm* 60 mm* 1 mm.

SEM Characterization: The microstructures of freeze-dried UCST-LCST hydrogels were analyzed by SEM S4800. The samples were frozen in liquid nitrogen for 10 min before lyophilizing with a freeze drier (FD-1C-50, Beijing BoYiKang) at −35 °C for about 24 h.

Thermal-responsiveness Characterization: The transmittance measurements were performed on a Cary 100 UV/Vis-Spectrophotometer equipped with a temperature controller. The single layer hydrogels were fabricated in situ in a quartz cell with 1 mm optical path. The wavelength was set at 600 nm, and the samples were heated from 25°C to 50°C at a constant heating rate of 1°C /min. UCST and LCST were determined as the temperatures at 50% transmittance respectively.

For bending test, two glass dishes loaded with liquid paraffin were heated to 15°C and 40°C, respectively, and a 20 mm* 5 mm* 2 mm bilayer hydrogel stripe was put into a 15°C dish for 30 min until it reached the equilibrium state. Then the stripe was then transferred into a 40°C dish. The stripe bended quickly toward the PNIPAM side in response to the elevated temperature. Once the curvature approached 360°, the stripe was transferred to the 15°C dish again to recover for 1 h. The bending-recovery experiment was repeated several times to determine the cycle performance. The process was recorded with a digital camera allowing a further study. The same tests were conducted under water instead of liquid paraffin.

The bending angles were measured according to Fig. S3.

In order to observe the change of the thickness of each layer clearly, the bilayer strips used in the thickness-changing test were 20 mm*1 mm*2 mm.

Fabrication of hydrogel gripper: The flower shape bilayer hydrogel was prepared by using a flower shape silica rubber spacer with a diameter of 1.5 cm. The hydrogel griper was fabricated by penetrating the hydrogel flower with a cotton fiber.

Supplementary Figures

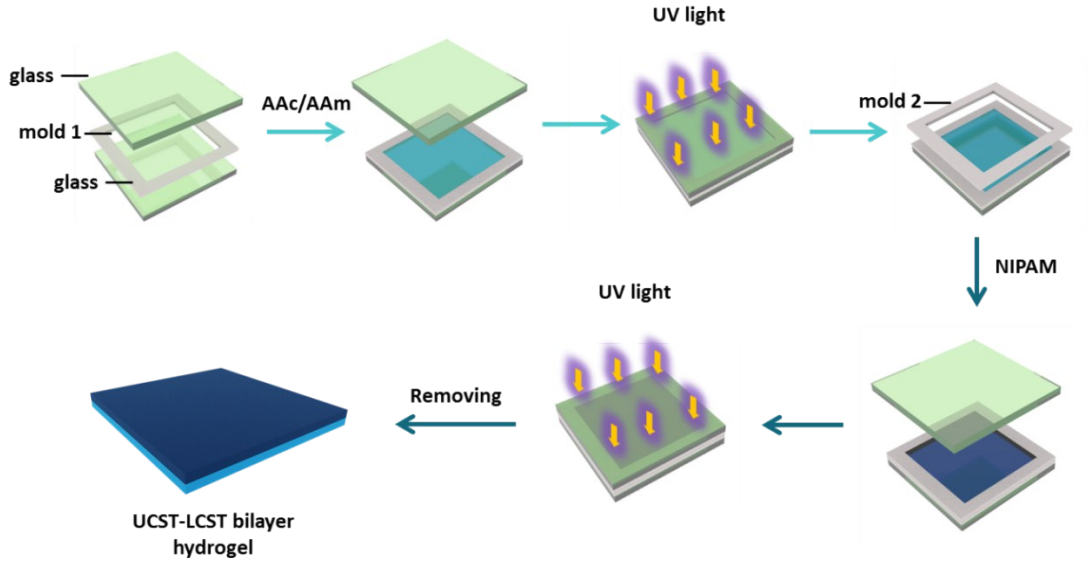

Fig. S1 Synthetic scheme of the UCST-LCST bilayer hydrogel.

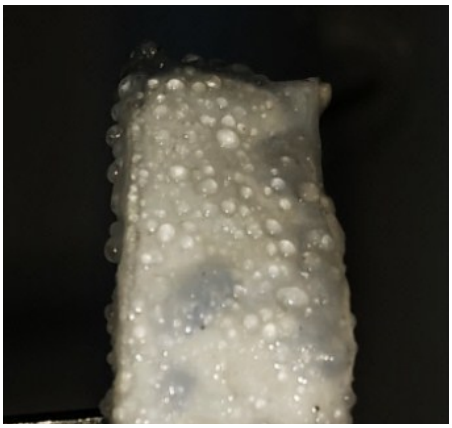

Fig. S2 Optical picture of bubble-like structures on the hydrogel surface formed during the heating process if no PEG was added during the bilayer hydrogel formation.

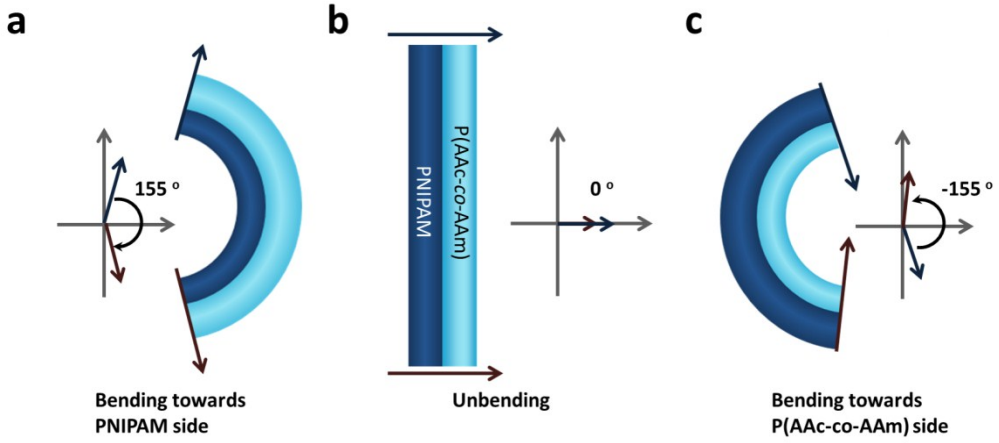

Fig. S3 The measurement of the bending angles. Bending towards the PNIPAM layer is marked as a positive angle while towards the P(AAm-*co*-AAc) layer is marked as a negative angle.

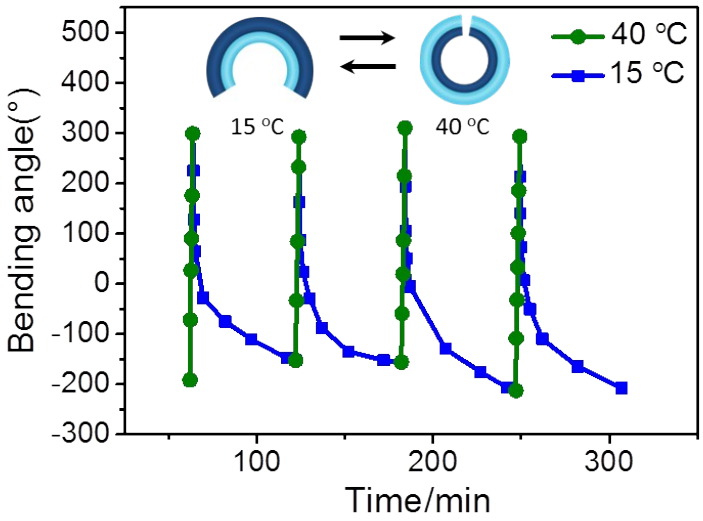

Fig. S4 Cyclic bending-unbending between 40°C and 15°C of the bilayer hydrogel in water.

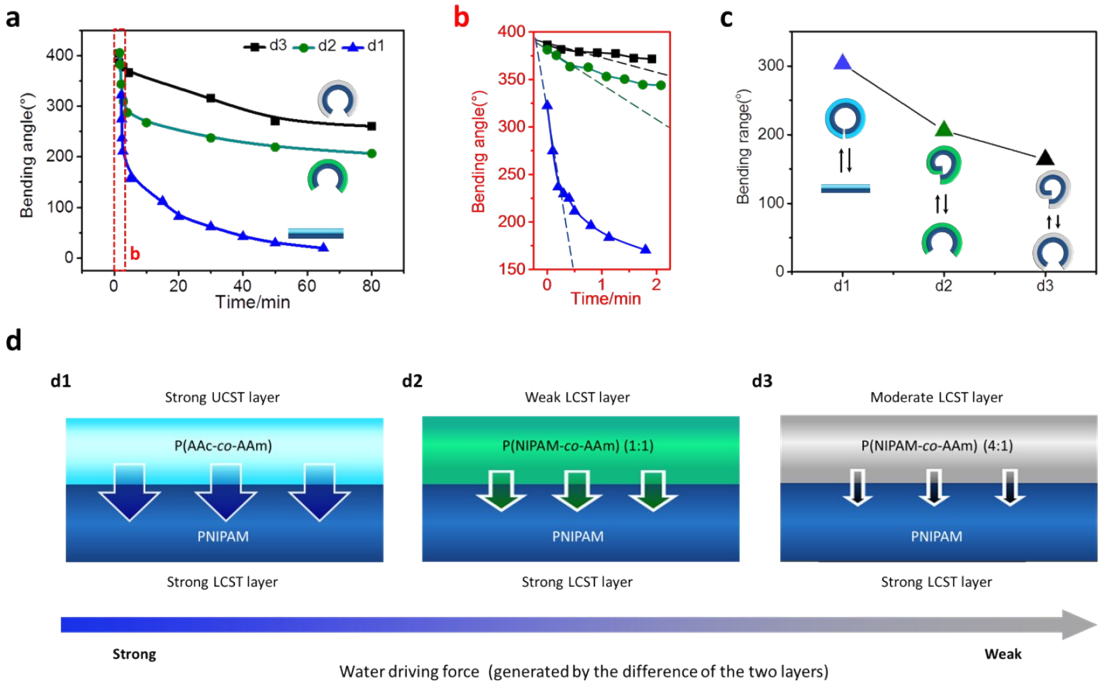

Fig. S5 (a-b) Changes of bending angle for different bilayer hydrogels when changing temperature from 40°C to 15°C in liquid paraffin. (c) The difference of the bending angles for different bilayer hydrogels when changing temperature from 40°C to 15°C. (d) Schematics of different bilayer hydrogels with varying polymer layers on top of a PNIPAM layer, resulting in varying driving forces for the self-circulation water transfer between the two layers.

Supplementary Movies

Movie S1 shows the responsiveness of a mimosa leaf.

Movie S2 shows the responsiveness of a mimosa petiole.

Movie S3 shows the closing and reopening of a flower shape bilayer hydrogel in water.

Movie S4 shows the closing and reopening of a flower shape bilayer hydrogel in liquid paraffin.

Movie S5 shows the closing and reopening of a flower shape bilayer hydrogel in open-air.

Movie S6 shows the lifting of porcelain balls in hot and cold environment by a hydrogel gripper.

Movie S7 shows the transferring of a cargo by a hydrogel gripper.