Supporting information

Bilayer hydrogel mixed composites that respond to multiple stimuli for environmental sensing and underwater actuation

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1. Fabrication of two types of hydrogels

Two types of hydrogels were fabricated through two-step polymerization. As to the "N" type, PNIPAM-AM layer consisted of NIPAM and AM with the molar ratio of 0.94:1. As to the "D" type, the PDMAEMA-AM layer consisted of DMAEMA and AM with the molar ratio of 0.3:1. Besides monomers, each layer of hydrogel consists of alginate and crosslinker PEGDA. Alginate polymer and monomer (the weight ratio of alginate to monomer was 0.03:1) were dissolved in deionized water, in which the combined amount of alginate and monomer was 25 wt%. The concentration of PEGDA in PNIPAM-only layer, PNIPAM-AM layer, PDMAEMA-only layer and PDMAEMA-AM layer maintained as 0.18 mol%, 0.45 mol%, 1.83 mol% and 1.15 mol%, respectively, relative to combined amount of monomers. Dyes in two types of hydrogels

maintained with the concentration of 1.09×10^{-3} mM.

2. Calculation of bending curvature (*k*)

The curvature during oriented deformation of bilayer hydrogel can be calculated with a facile method. Concerning that bending towards each layer is caused by shrinkage or swelling of one layer, the length of another one can be treated as unchanged. As shown in Fig. S1, the radius of curvature and film-free length are regarded as r and L; thus curvature (*k*) corresponds to 1/r. During the measurement, the bending behavior is uniform on each position of the bilayer hydrogel, suggesting that the equation **(2)** of curvature calculation is adaptable for this system. Through measuring the deflection angle and film-free length, the curvature variation can be obtained.

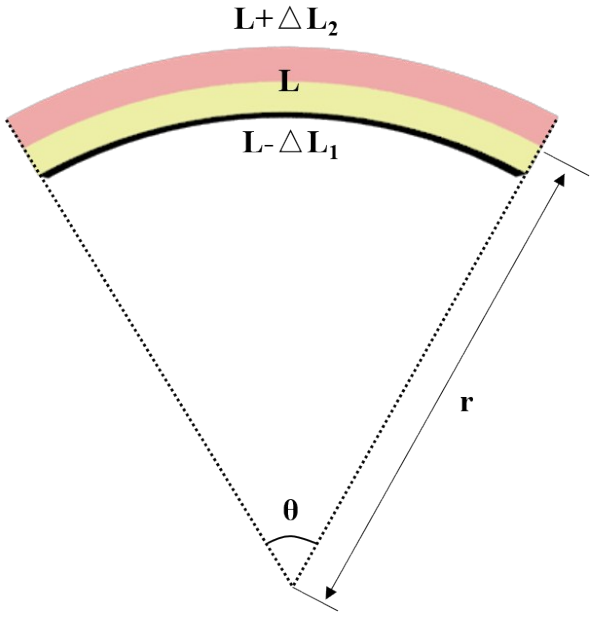

Fig. S1. Scheme representation of bending behavior analysis of bilayer hydrogel. θ is the bending angle, L is the length of the film, ΔL is the length change between two layers

3. Temperature response of "N" type hydrogel

Percentage area loss (AL%) of two layers is proposed to compare the shrinkage

difference within varied temperature as follows:

$$AL\% = \frac{(S_0 - S_1)}{S_0} \times 100\% \quad (1)$$

where S_1 is the area of hydrogel in hot water, S_0 is the area of prepared hydrogel.

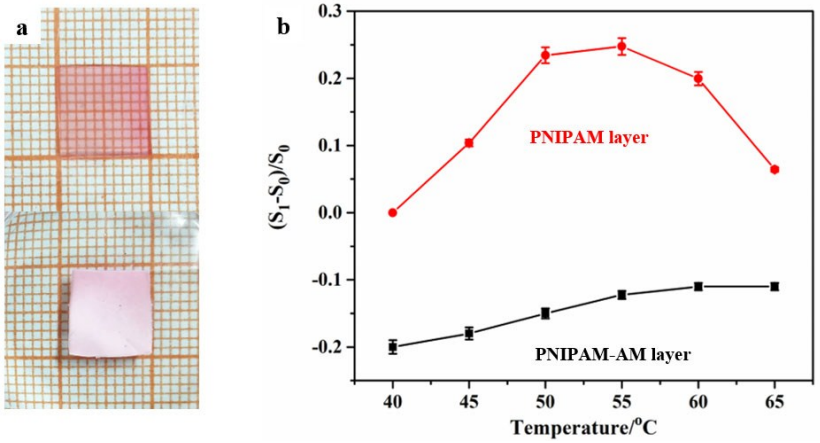

Fig. S2 T response of PNIPAM/PNIPAM-AM hydrogel. (a) Volume change of PNIPAM hydrogel in hot water (45 °C). (b) The relative area changes of two layers at different temperatures.

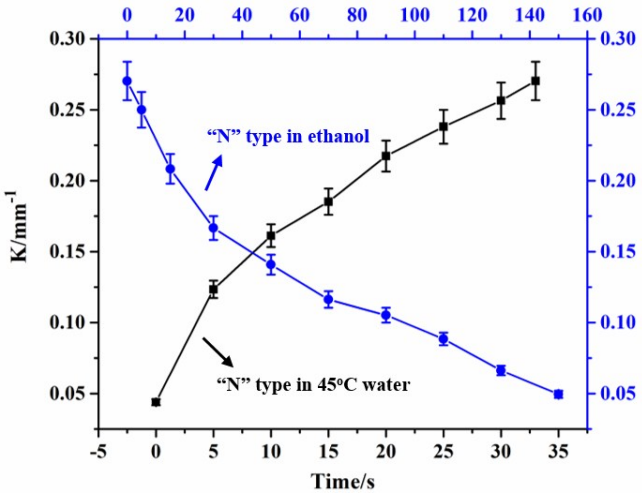

Fig. S3 Curvature variation of "N" type hydrogel in hot water and ethanol successively.

4. Temperature response of "D" type hydrogel

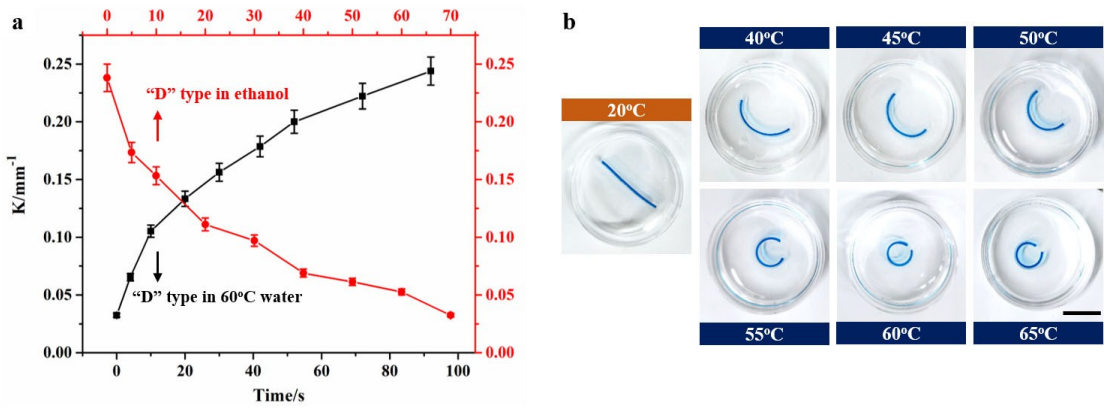

Fig. S4 (a) Curvature variation of "D" type hydrogel in hot water and ethanol successively. (b) The initial state and deformation state of "D" type hydrogel at different temperature for 60 s. The scale bar is 1 cm.

5. Stimuli-responsive properties of "N" and "D" type hydrogels

Both of the two types hydrogels possess dual-response towards environmental stimuli. As to "N" type, it is sensitive towards temperature and salt, suggesting bending deformation towards PNIPAM layer in hot water and NaCl solution. The recovery of such bending can be realized by immersing into ethanol. While in "D" type, pH and temperature can influence the swelling of hydrogel and cause bending towards PDMAEMA-AM (acid) and PDMAEMA (hot water) layer respectively. Table S1 illustrates bending directions of two types of hydrogels in different environments.

Table S1. Bending directions of two types of hydrogels in different environments (hot water/acid/salt solution and ethanol)

	hot water	acid	salt solution	ethanol
PNIPAM layer	+	**none**	+	
PNIPAM-AM layer				**+ (slight)**
PDMAEMA layer	+		**none**	
PDMAEMA-AM layer		+		**+ (distinct)**

"+" represents the bending towards layer

6. Hydrogel composites

Given the pre-programmed design of hydrogel integration, we can obtain various

motions through combination of “N” and “D” types. We glue the segments together and obtain the original linear state, as shown in Figure S5. The composition and treatment of each composites have been proposed.

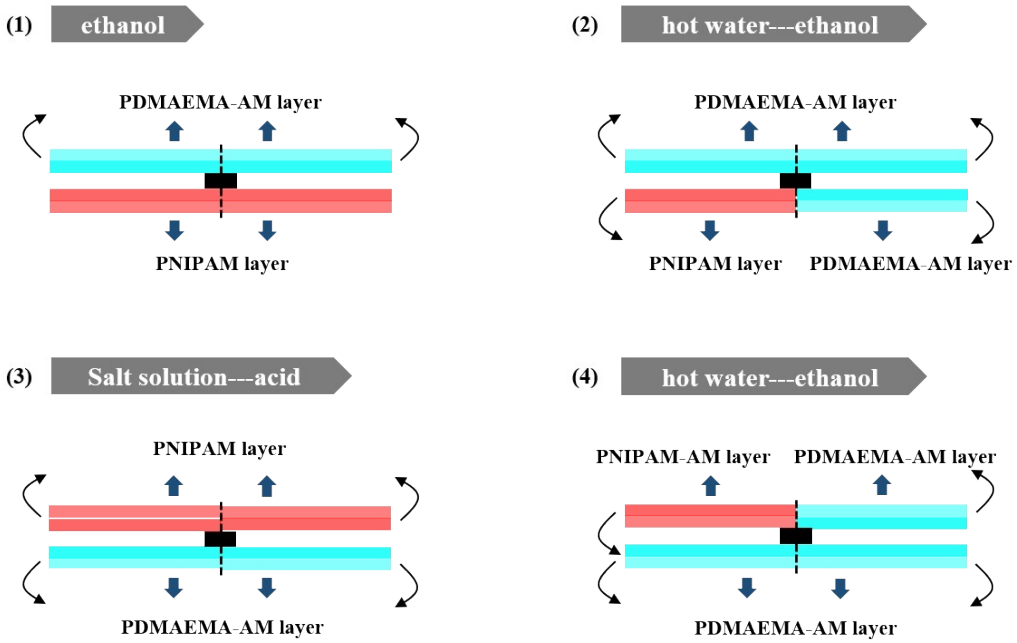

Fig. S5 Schematic illustration of hydrogel composites composition and treatment in Figure 8c. (1)-(4) correspond to the four motions in Figure 8c.