Supporting Information

A monolithic anti-freezing hydro/organo Janus actuator with sensitivity to polarity of solvents

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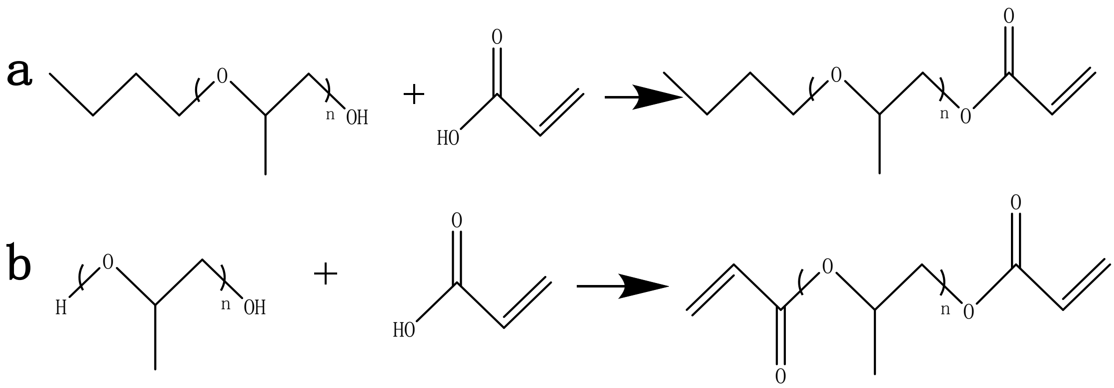
1. Synthesis and Structure Characterization of PPGMA and PPGDA



Scheme S1. Synthetic Routes of (a) PPGMA and (b) PPGDA.

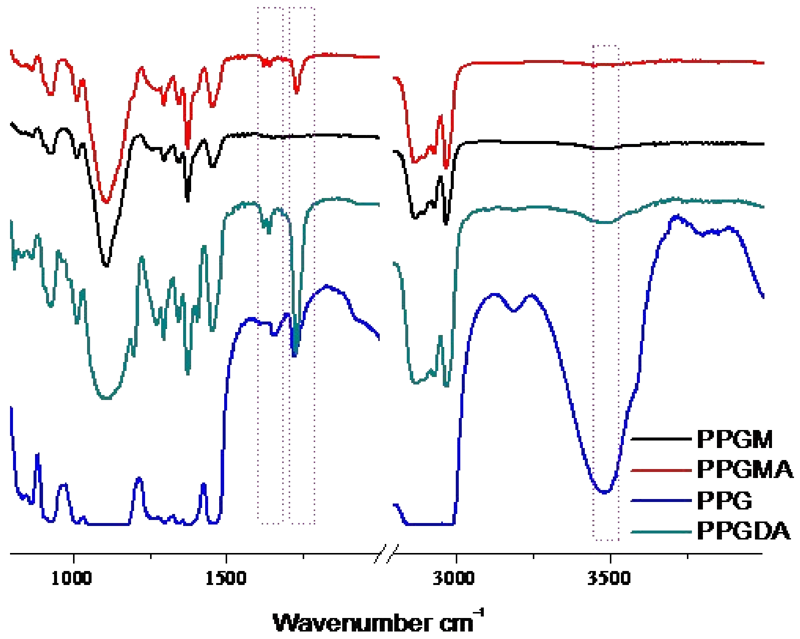


Figure S1. FTIR spectra of PPGM , PPGMA , PPG and PPGDA.

S. No.	Wave no.range	Peaks in PPG	Peaks in PPGDA	Peaks in PPGM	Peaks in PPGMA	Assignment
1	3500-3700	3480	Low intensity	3473	Low intensity	-OH Str. in alcohols
2	3000-2840	2844-2989	2879，2966	2865,2969	2868,2973	-CH Str. in alkanes
3	1680-1760	Low intensity	1722	Absent	1727	-C = O Str. for easter group conjugated with –C = C-
4	1580-1630	Low intensity	1635	Low intensity	1621	-C = C- Str. in acrylate bond
5	1370-1485	1305,1402	1405,1459	1374,1457	1375,1457	-CH def. in methyl/ methylene
6	1050-1300	Low intensity	1195	1109	1109	C-O stretching

Table S1 . FTIR Peaks of synthesized monomers[1].

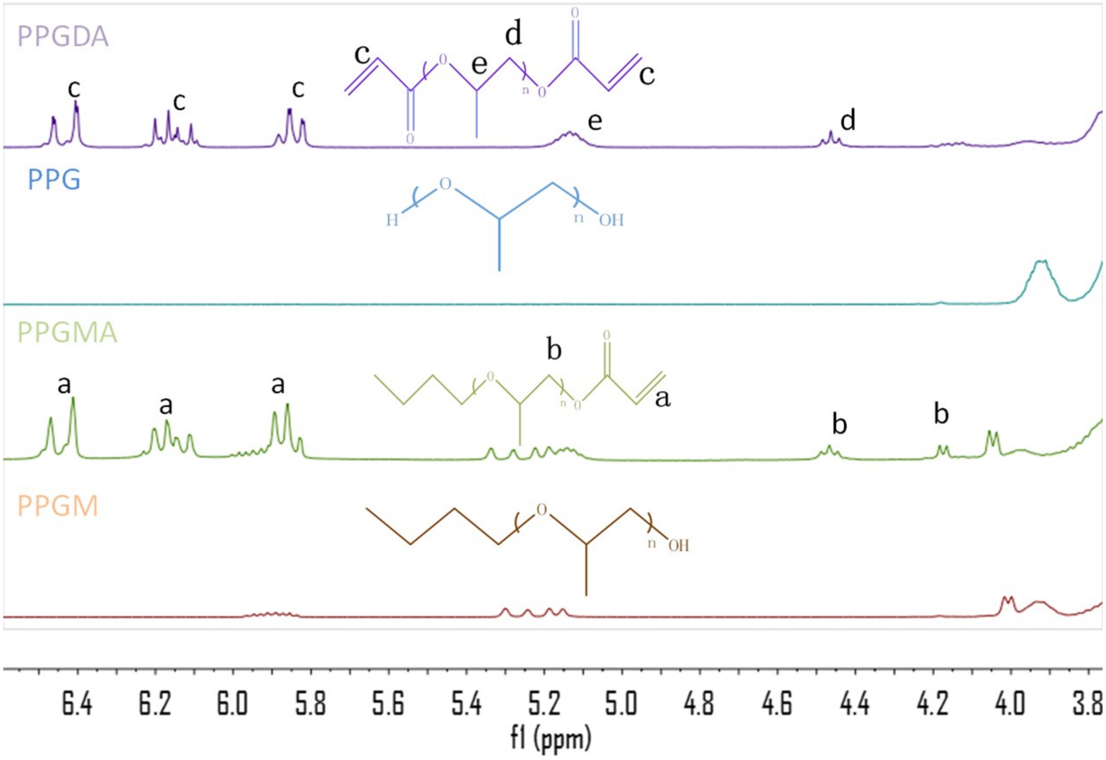
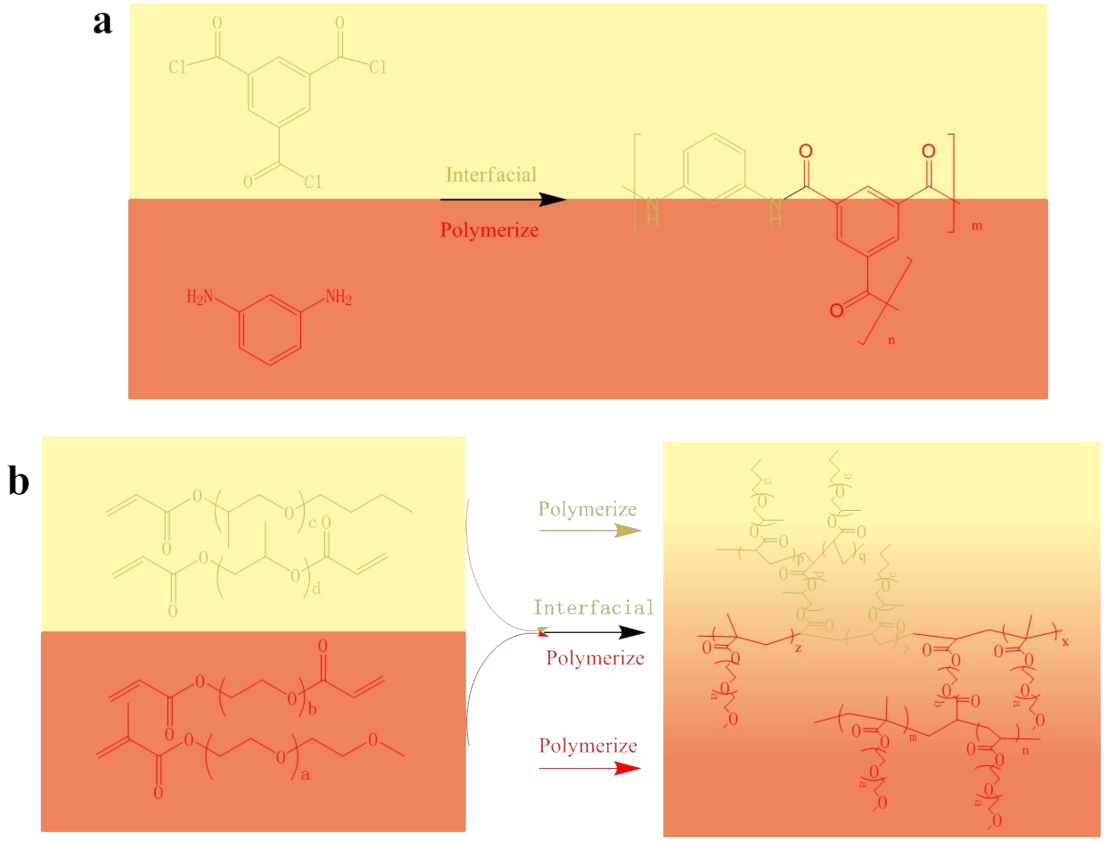


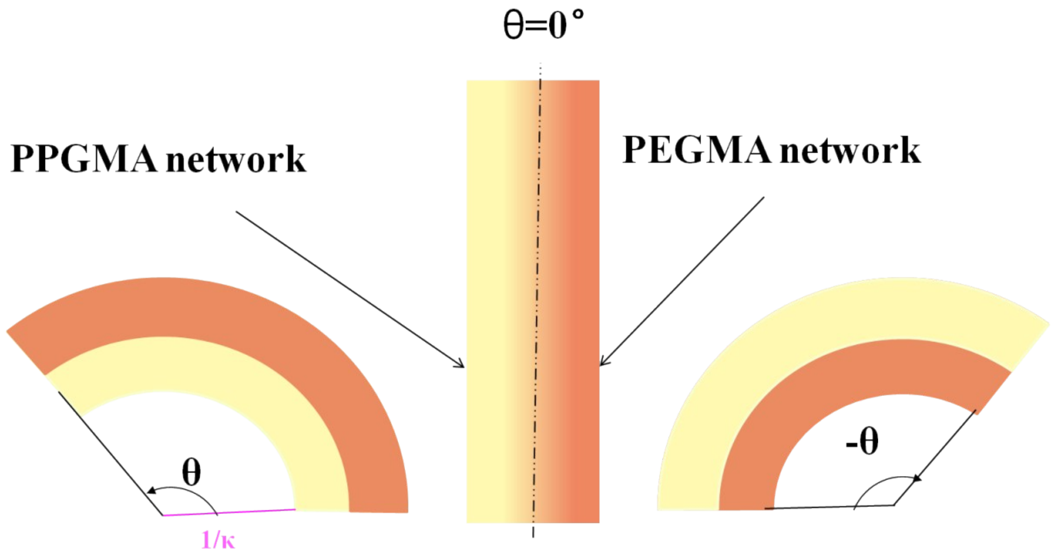
Figure S2. ¹H-NMR spectra and structural illustrations of PPGDA, PPG, PPGMA and PPGM.

2. **The unconventional interfacial polymerization mechanism to for fabricating hydro/organo macro copolymer Janus strip[2].**



Scheme S2.The differences between traditional interface polymerization and our interface copolymerization. (A) The conventional interfacial polymerization is actually the polycondensation reaction of polyamine and polyacid at the oil / water interface. (B) Our interfacial copolymerization is actually a free radical polymerization reaction of hydrophilic vinyl monomers and hydrophobic vinyl monomers at the oil / water interface.

Our hydro/organo janus copolymer strip was obtained by interfacial copolymerization. The unified Janus strip was prepared through one-step interface copolymerization with hydro network and organ network. This interfacial copolymerization, in contrast to conventional interfacial polymerization (condensation of oil / water interfaces) (Scheme S3.a), is an additional polymerization of hydrophilic vinyl monomers with hydrophobic vinyl monomers at the oil-water interface (Scheme S3.b). Due to the interfacial copolymerization, the organogel network and the hydrogel network can be well connected by the interface network.



Scheme S3. The definition of bending angle. A negative angle (-θ) represents bending inward to the PEGMA network side of the Janus strip, whereas a positive angle (θ) represents bending inward to the PPGMA network of the Janus strip.

3. Janus properties and actuation performance of the PEGMA-B-PPGMA strip.

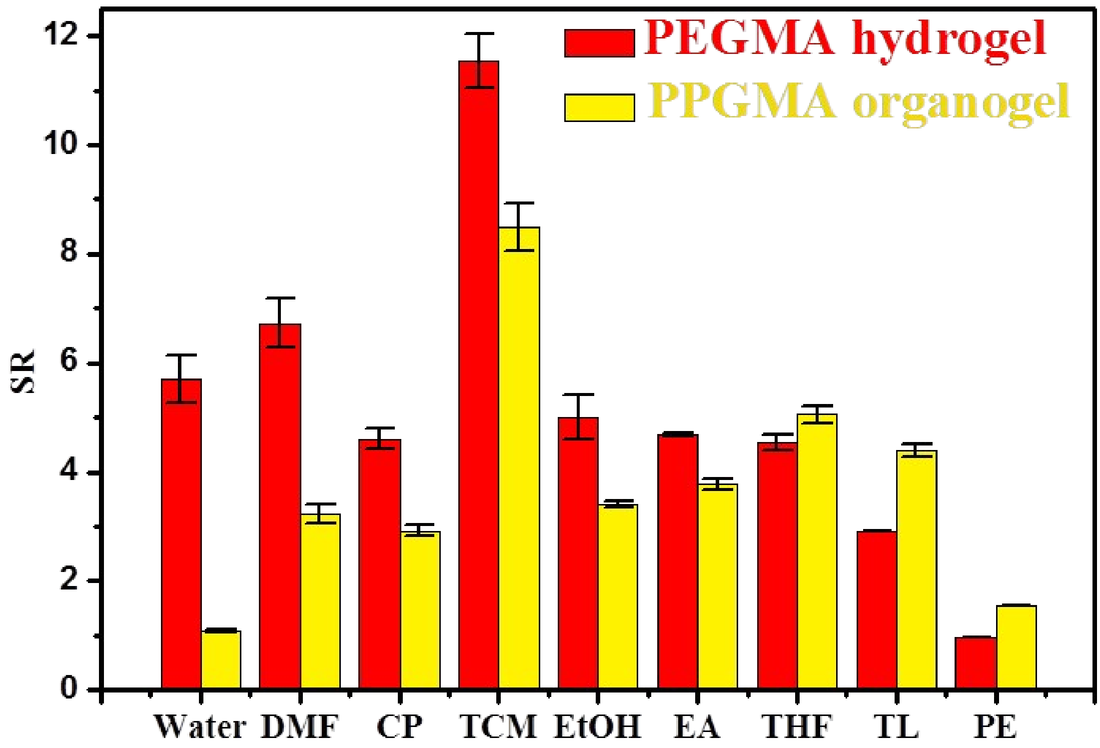


Figure S3. The swelling ratio of pure PEGMA hydrogel and pure PPGMA organogel in various solvents.

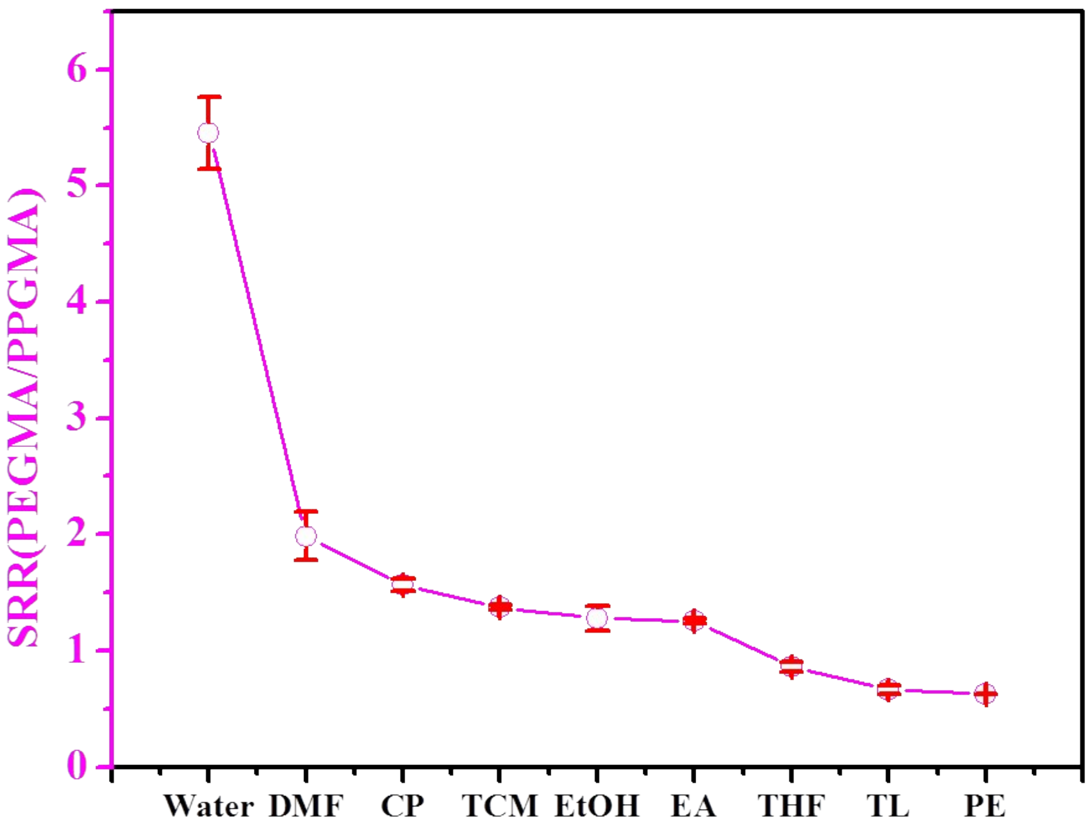


Figure S4. Relationship between SRR(PEGMA/PPGMA) and solvent polarity.

With the decrease of the polarity of organic solvents,

SRR=SR(PEGMA)/SR(PPGMA) also showed a decreasing trend.

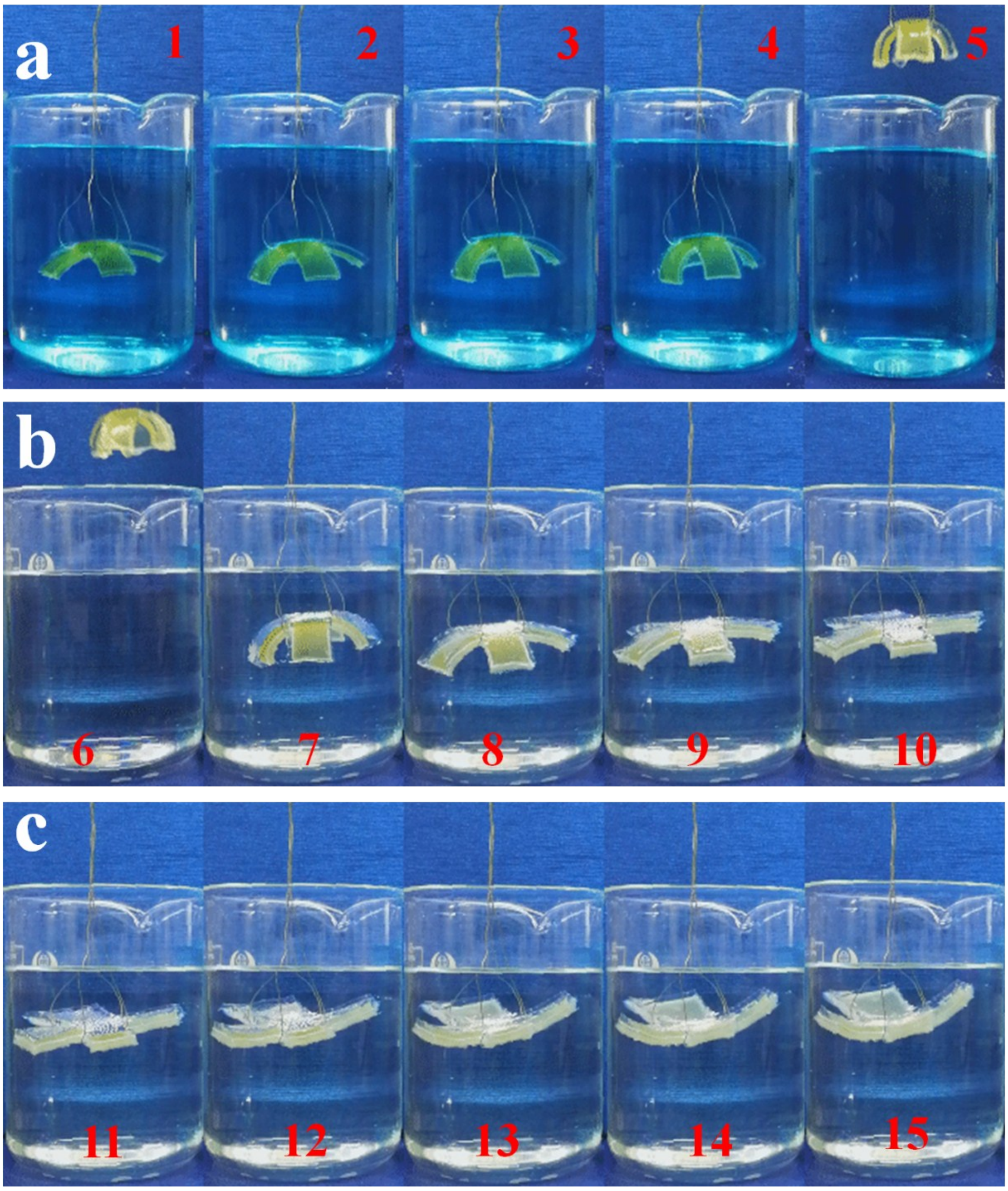


Figure S5. Photos of the reversible bending of the PEGMA-B-PPGMA Janus strip in aqueous solution and toluene. The "+" shaped PEGMA-B-PPGMA Janus strip, (a) first placed in a beaker containing toluene, gradually bent downward(1→4). (b)Then removed and placed in a beaker containing water(5→6), it gradually bent upward, (c) eventually lead to upward bending(7→15).

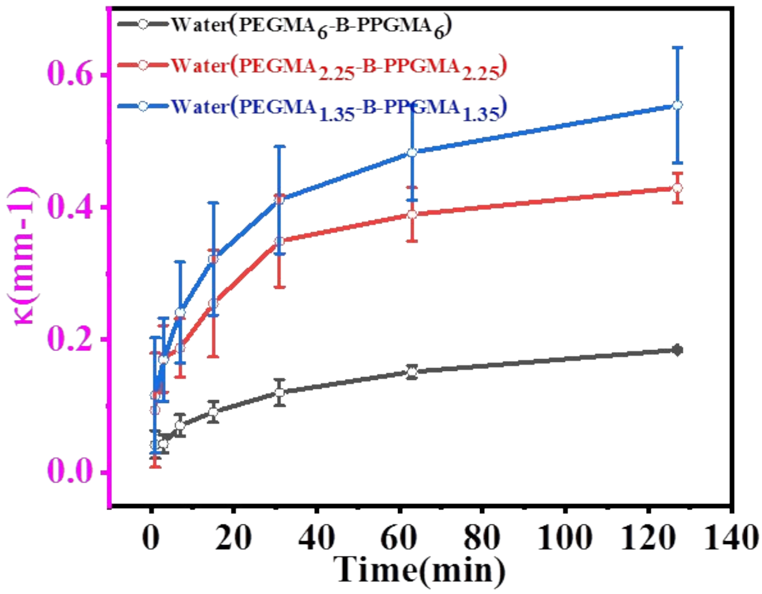


Figure S6. Corresponding curvature of the PEGMA$_x$-B-PPGMA$_y$ (Where x and y respectively represents the volume of water-soluble solution and organic solution added to the mold) Janus actuator strips during the dynamic process in water. When V_{PEGMA}/V_{PPGMA} is constant (V_{PEGMA}/V_{PPGMA}=1), the curvature decreases with the increase of total thickness, the response time can be reduce by decreasing thickness of samples.

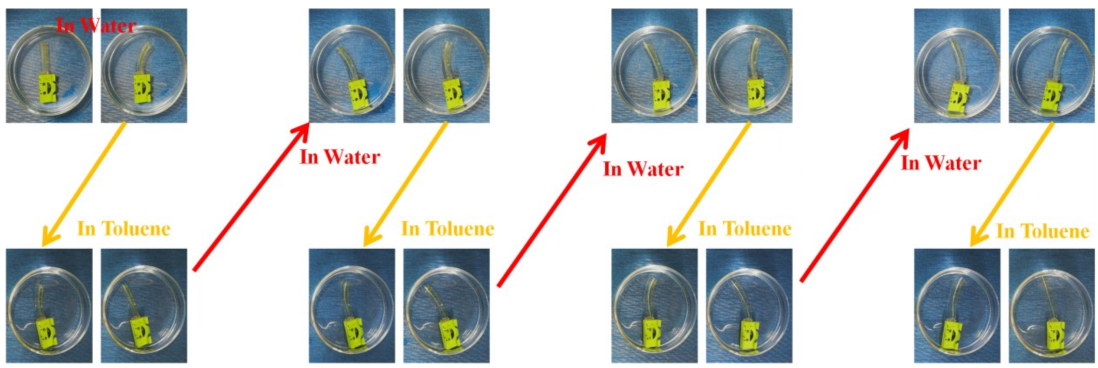


Figure S7. Photos of the reversible bending of the PEGMA-B-PPGMA Janus strip in aqueous solution and toluene. All photos from the screenshot of Movie S1

Figure S8. **Bending photographs of the PEGMA-B-PPGMA Janus copolymer strip（frozen at -20°C for 24 h）in toluene at -20°C.**

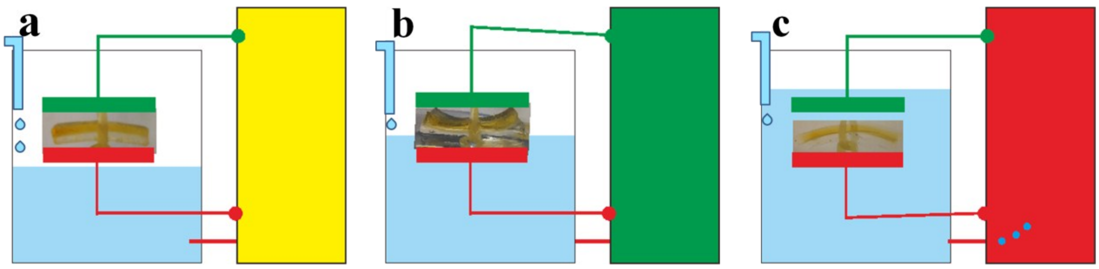


Figure S9. PEGMA-B-PPGMA Janus strip Application principle schematic (a) The leaked solvent did not reach the warning line (b) The leaked solvent reached a warning line (first response) (c) The leaked solvent reached the second warning line (second response).

4. Supplementary Movies

Movie S1. *In-situ* observation of the reversible bending of the PEGMA-B-PPGMA Janus strip in aqueous solution and toluene with a digital camera. The video was played at a 64× speed.

Movie S2. *In-situ* observation of the reversible bending of the "+" shaped PAA-B-PBMA Janus strip in toluene and water with a digital camera. The video was played at a 48× speed.

Movie S3. Device for organic solvents leakage warning with the PEGMA/PPGMA Janus strip as the sensor element. When high polar solvents (DMF as simulant solvent) leak, the Janus strip bent towards right (the PPGMA network inward), closed the right circuit and then lightened the green lamp.

Movie S4. In-situ observation of the twice response of the PEGMA-B-PPGMA Janus strip in ethyl acetate with a digital camera. When less than half of the Janus strip was immersed in the solvent（ethyl acetate）, the Janus strip was found to bend upwards and produce the first response. When the Janus strip was completely immersed in the solvent, the second response occured and the Janus strip gradually flattened or even bent downwards. The video was played at a 16× speed.

1. S. Shukla and J. S. P. Rai, *International Journal of Plastics Technology*, 2013, **17**, 182-193.

2. F. Zhang, J. Fan, P. Zhang, M. Liu, J. Meng, L. Jiang and S. Wang, *Npg Asia Materials*, 2017, **9**， e380.