Supporting Information:

Spatially and Reversibly Actuating Soft Gel Structure by Harnessing Multimode Elastic Instabilities

Yingzhi Liu^{1,2§}, Ansu Sun^{2§}, Sreepathy Sridhar², Zhenghong Li^{1,2}, Zhuofan Qin², Ji Liu³, Xue Chen², Haibao

 Lu^{1*} , Ben Zhong Tang^{4*} and Ben Bin Xu^{2*}

¹Science and Technology on Advanced Composites in Special Environments Laboratory, Harbin Institute of Technology, Harbin, Heilongjiang, 150080, China.

²Smart Materials and Surfaces Laboratory, Faculty of Engineering and Environment, Northumbria University, Newcastle upon Tyne, NE1 8ST, UK.

³Department of Mechanical and Energy Engineering, Southern University of Science and Technology, Shenzhen, 518055, China.

⁴Department of Chemistry, The Hong Kong Branch of Chinese National Engineering Research Center for Tissue Restoration and Reconstruction and Institute for Advanced Study, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, 999077, Hong Kong, China.

*Corresponding Author:

E-mail: <u>luhb@hit.edu.cn</u> (Haibao Lu);

E-mail: tangbenz@ust.hk (Ben Zhong Tang);

E-mail: <u>ben.xu@northumbria.ac.uk</u> (Ben Bin Xu)

[§]Yingzhi Liu and Ansu Sun contributed equally to this work.

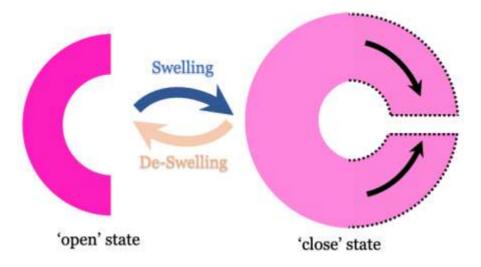


Figure S1. Schematic of reversible morphing of 'semi-cylinder shell' shaped gel part under swelling/de-swelling.

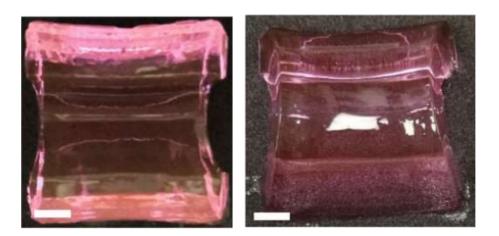


Figure S2. Final equilibrium state of gel structure at an immersion time of 240 min. Observations from the axial edges (left) and circumferential out surface (right). The scale bar is 5 mm.

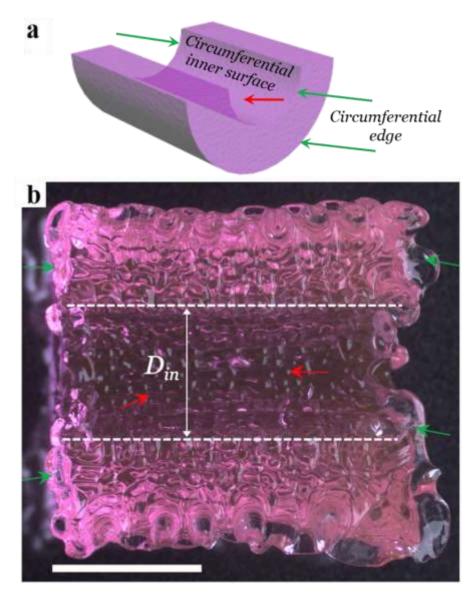


Figure S3. Buckling on circumferential edge and inner zone. (a) The schematic of the circumferential edge (green arrow) and the circumferential inner zone (red arrow). (b) Optical micrograph of gel structure swelling at 2 mins, the dash lines highlight the inner edge with a label D_{in} . The gel structure has initial $D_{out}=L_0=10 \text{ mm}$, $D_{in}=5 \text{ mm}$, $\theta=180^\circ$. The scale bar is 5 mm.

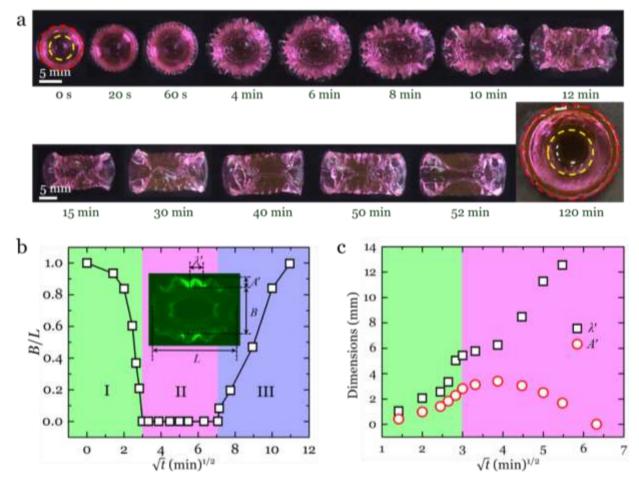


Figure S4. Swelling-induced mechanical instabilities of the hemispherical shell hydrogel. (a) The observations of morphologies at various time intervals of the hemispherical shell hydrogel under optical microscope. The analytical plots of time dependent evolutions of structure deformation during swelling: (b) B/L, where B refers to the gap distance between edges, L is defined as the structure length of swollen hydrogel, green area: I-closing regime, pink area: II-holding regime, purple area: III-reopening regime; (c) wavelength (λ') and amplitude (A') of buckling.

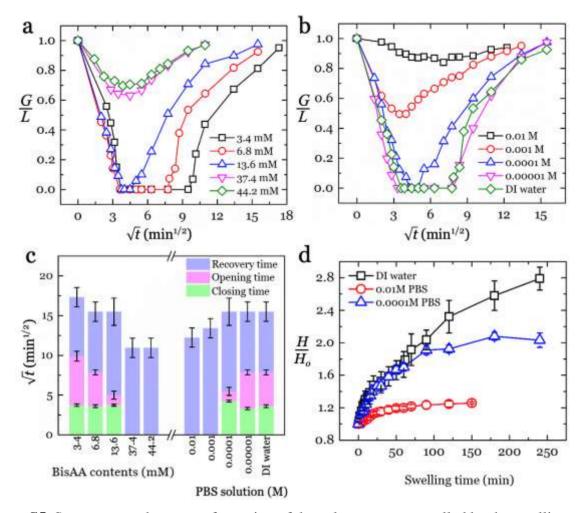


Figure S5. Spontaneous shape transformation of the gel structure controlled by the swelling ratio. Time evolution of the shape deformation of gel structure with initial $D_{out}=L_0=10$ mm, $D_{in}=5$ mm, $\theta=180^{\circ}$: (a) with different BisAA contents swelling in DI water, (b) swelling in PBS with different concentrations. (c) The influence of crosslinking and solution on shape deformation times of gel structure. (d) The swelling ratio as function of time in different PBS.

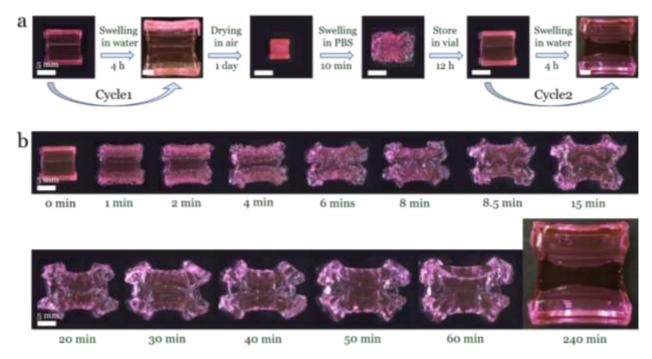


Figure S6. Reproducibility of the actuating gel structure. (a) The experiment design. (b) Time evolution of shape deformation of gel structure with initial $D_{out}=L_0=10$ mm, $D_{in}=5$ mm, $\theta=180^{\circ}$ during cycle 2.

Supplementary Movie S1. Development of the wavy buckling profile on the axial edges by an optical microscope.

Supplementary Movie S2. Finite element analysis results.

Supplementary Movie S3. Demonstration of gripping strength using a designed hydrogel

gripper to pick up a magnetic stirrer from water.