

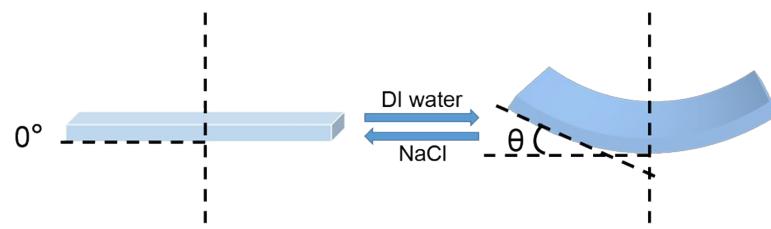
## Supporting Information

# Salt-Responsive Polyampholyte-based Hydrogel Actuators with Gradient Porous Structure

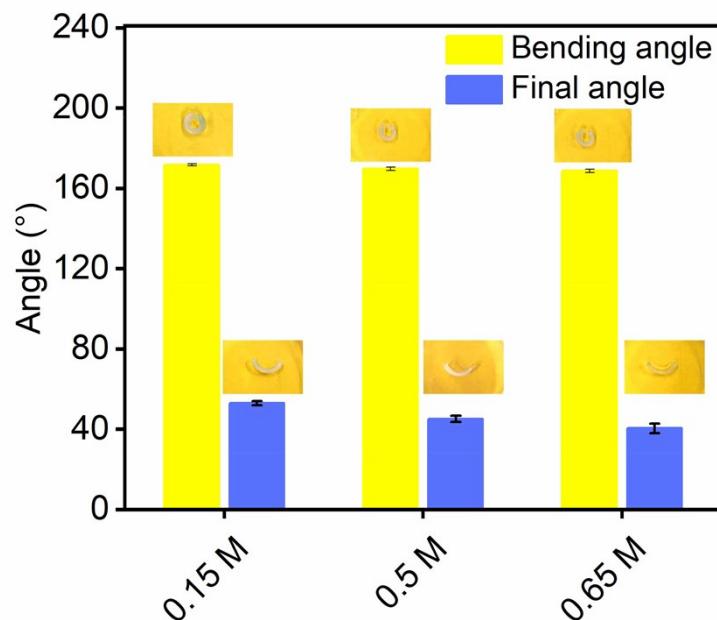
## Experimental section

### Synthesis of 2-ureidoethyl methacrylate

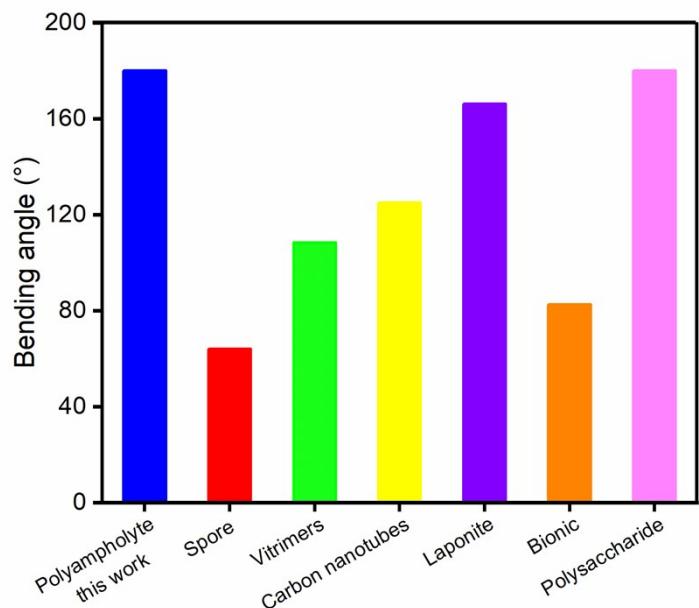
2-ureidoethyl methacrylate (UM) was synthesized according to a previous work.<sup>1</sup> Briefly, 30% aqueous solution of ammonia (7.8 g) was added into a three-necked flask with a dropping funnel, reflux condenser and a stirrer. The solution was cooled to 10 °C and 7.75 g 2-isocyanatoethyl methacrylate was added. Along with the reaction proceeded, white crystals precipitated and the crystals was washed with acetone and dried under vacuum. 8.5 g UM (white powder, yield: 90%) was finally obtained.



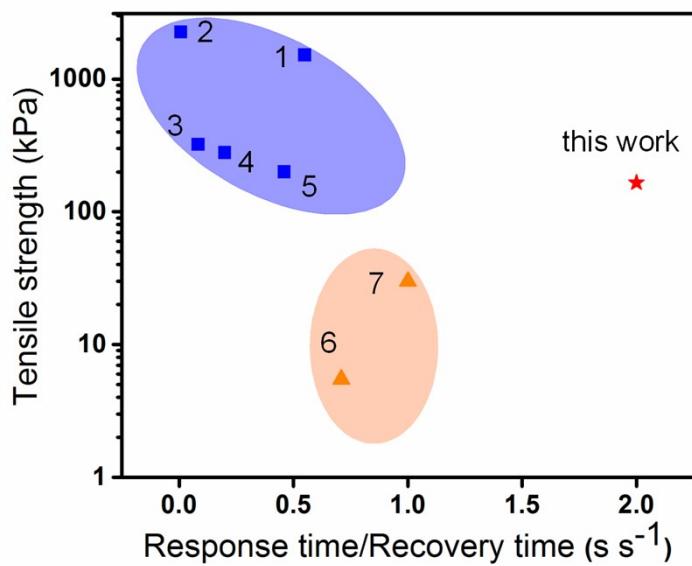
**Figure S1.** Definition of bending angle and final angle. The angle of the strip in the sodium chloride solution is defined as  $0^\circ$ .



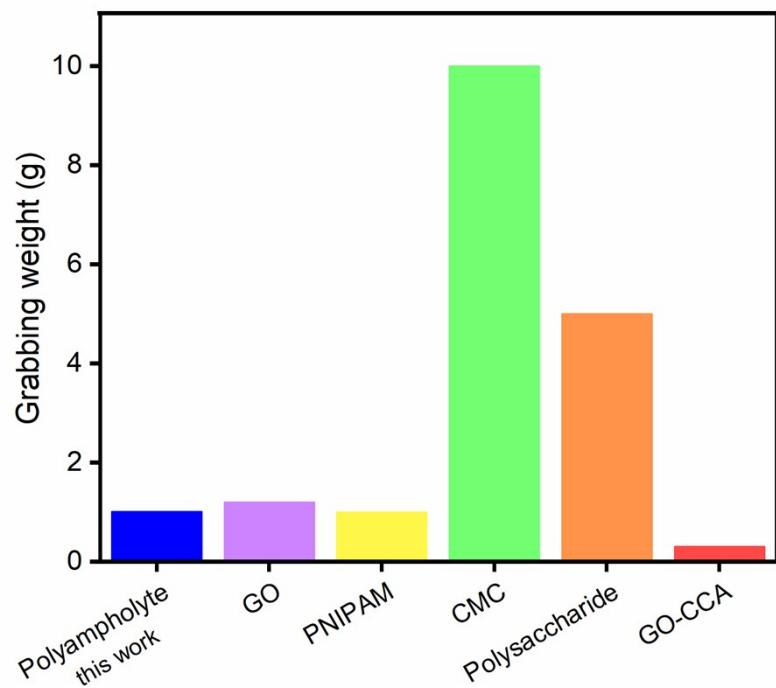
**Figure S2.** The bending angle and final angle of actuators in the NaCl solutions with low concentrations (0.15 M and 0.65 M NaCl represent normal saline and seawater, respectively).



**Figure S3.** Comparison of bending angle between the hydrogel actuator and other systems, the details are shown in Table S1.



**Figure S4.** Comparison of tensile strength and response/recovery time ratio between the hydrogel actuator and other hydrogel actuators (No. 1-7).



**Figure S5.** Comparison of grabbing weight between the polyampholyte-based gripper and GO<sup>14</sup>, PNIPAM<sup>15</sup>, CMC<sup>16</sup>, Polysaccharide<sup>17</sup>, GO-CCA<sup>18</sup> systems.

**Table S1.** Data collection for Figure S3. Hydrogel actuators with different bending angle, response time and bending rate are considered.

Materials	Bending angle (°)	Response time (s)	Bending rate (°/s)	Ref.
Polyampholyte	180.0	3600	0.05	This work
Spore	64.1	40	1.60	<sup>2</sup>
Vitrimers	108.4	600	0.18	<sup>3</sup>
Carbon nanotubes	125.0	4800	0.02	<sup>4</sup>
Laponite	166.3	120	1.38	<sup>5</sup>
Bionic	82.6	480	0.17	<sup>6</sup>
Polysaccharide	180.0	15	12	<sup>7</sup>

**Table S2.** Data collection for Figure S4. Hydrogel actuators with different tensile strength and response/recovery time ratio are considered.

Materials	Tensile strength (kPa)	Response/Recover y time ratio	Number	Ref.
P(NaSS- <i>co</i> -DMAEA-Q)	165	2	This work	This work
P(AAc- <i>co</i> -NIPAm)	1520	$5.5 \times 10^{-1}$	1	8
CS/SA	2260	$8.0 \times 10^{-3}$	2	7
Al-alginate/PNIPAM	322	$8.3 \times 10^{-2}$	3	9
PNIPAM/Laponite	280	$2.0 \times 10^{-1}$	4	10
TiO <sub>2</sub> /AM/NIPAM	200	$4.6 \times 10^{-1}$	5	11
P(Fc- <i>co</i> - $\beta$ -CD-AAm)	6	$7.1 \times 10^{-1}$	6	12
P(NIPAM)/P(VBIPS)	30	1	7	13

**Movie S1.** The shape deformation process of the gradient P(NaSS-*co*-DMAEA-Q) hydrogel actuator in deionized (DI) water.

**Movie S2.** The shape recovery process of the gradient P(NaSS-*co*-DMAEA-Q) hydrogel actuator in NaCl solution.

**Movie S3.** The grabbing-releasing process of the underwater gripper in DI water and NaCl solution.

## References

1. *Japan Pat.*, JP2005104879, 2005.
2. O. Cakmak, H. O. El Tinay, X. Chen and O. Sahin, *Adv. Mater. Technol.*, 2019, **4**.
3. Q. Chen, X. Qian, Y. Xu, Y. Yang, Y. Wei and Y. Ji, *ACS Appl. Mater. Interfaces*, 2019, **11**, 29290.
4. A. Toncheva, B. Willocq, F. Khelifa, O. Douheret, P. Lambert, P. Dubois and J.-M. Raquez, *J. Mater. Chem. B*, 2017, **5**, 5556.
5. Y. Tan, D. Wang, H. Xu, Y. Yang, X. Wang, F. Tian, P. Xu, W. An, X. Zhao and S. Xu, *ACS Appl. Mater. Interfaces*, 2018, **10**, 40125.
6. C. Ma, W. Lu, X. Yang, J. He, X. Le, L. Wang, J. Zhang, M. J. Serpe, Y. Huang and T. Chen, *Adv. Funct. Mater.*, 2018, **28**, 1704568.
7. H. Cui, N. Pan, W. Fan, C. Liu, Y. Li, Y. Xia and K. Sui, *Adv. Funct. Mater.*, 2019, **29**, 1807692.
8. S. Y. Zheng, Y. Shen, F. Zhu, J. Yin, J. Qian, J. Fu, Z. L. Wu and Q. Zheng, *Adv. Funct. Mater.*, 2018, **28**, 1803366.
9. W. J. Zheng, N. An, J. Yang, J. Zhou and Y. M. Chen, *ACS Appl. Mater. Interfaces*, 2015, **7**, 1758.
10. Y. Tan, D. Wang, H. Xu, Y. Yang, W. An, L. Yu, Z. Xiao and S. Xu, *Macromol. Rapid Commun.*, 2018, **39**, 1700863.
11. B. Xu, H. Jiang, H. Li, G. Zhang and Q. Zhang, *RSC Adv.*, 2015, **5**, 13167.
12. C. Ma, T. Li, Q. Zhao, X. Yang, J. Wu, Y. Luo and T. Xie, *Adv. Mater.*, 2014, **26**, 5665.
13. S. Xiao, M. Zhang, X. He, L. Huang, Y. Zhang, B. Ren, M. Zhong, Y. Chang, J. Yang and J. Zheng, *ACS Appl. Mater. Interfaces*, 2018, **10**, 21642.
14. T. Wang, J. Huang, Y. Yang, E. Zhang, W. Sun and Z. Tong, *ACS Appl. Mater. Interfaces*, 2015, **7**, 23423.
15. J. Zheng, P. Xiao, X. Le, W. Lu, P. Théato, C. Ma, B. Du, J. Zhang, Y. Huang and T. Chen, *J. Mater. Chem. C*, 2018, **6**, 1320.
16. S. Wu, F. Yu, H. Dong and X. Cao, *Carbohydr. Polym.*, 2017, **173**, 526.
17. J. Duan, X. Liang, K. Zhu, J. Guo and L. Zhang, *Soft Matter*, 2017, **13**, 345.
18. Z. Zhang, Z. Chen, Y. Wang, J. Chi, Y. Wang and Y. Zhao, *Small Methods*, 2019, **3**, 1900519.