## **Supporting Information**

## Nanoarchitectonics Composite Hydrogels with High Toughness, Mechanical

## Strength, and Self-Healing for Electrical Actuators with Programmable Shape

# **Memory Properties**

Yanqing Wang<sup>a</sup>, Pengcheng Li<sup>a</sup>, Shuting Cao<sup>a</sup>, Yuetao Liu<sup>a</sup>, Chuanhui Gao<sup>a\*</sup>

College of Chemical Engineering, Qingdao University of Science and Technology, Qingdao

266042, China.

\*Corresponding author: gaochuanhui@qust.edu.cn

Tel: +86 0532-84023170

Gly					
Content (g)	Mass fraction (wt%)	AAc (g)	Zr <sup>4+</sup> (g)	V-50 (g)	H <sub>2</sub> O (g)
0	0%	3.60	1.29	0.07	10.04
0.75	5%	3.60	1.29	0.07	9.29
1.50	10%	3.60	1.29	0.07	8.54
2.25	15%	3.60	1.29	0.07	7.79
3.00	20%	3.60	1.29	0.07	7.11

Table S1 The compositions of hydrogels with different glycerol concentrations.

The total volume of the precursor solution is kept as 15 mL.

PEI Mass Zr<sup>4+</sup> Content Gly V-50 AAc  $H_2O$ fraction **(g) (g) (g) (g) (g) (g)** (wt%) 7.64 0.15 1% 3.60 1.29 2.25 0.07 0.45 3% 3.60 1.29 2.25 0.07 7.34 0.75 7.04 5% 3.60 1.29 2.25 0.07 1.05 7% 3.60 1.29 2.25 0.07 6.74 1.35 9% 3.60 1.29 2.25 0.07 6.44 11% 4.90 1.50 3.60 1.29 2.25 0.07

**Table S2** The compositions of hydrogels with different PEI concentrations.

The total volume of the precursor solution is kept as 15 mL.

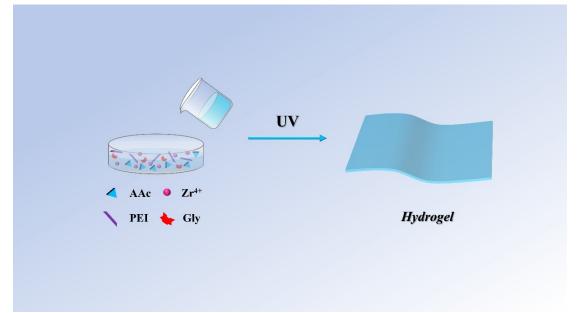


Figure S1. Diagram of the preparation process of the hydrogel.

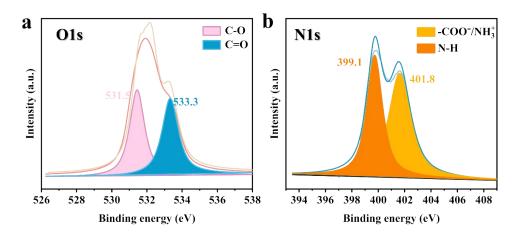


Figure S2. (a) O1s XPS spectra of PAEG hydrogel. (b) N1s XPS spectra of PAEG hydrogel.

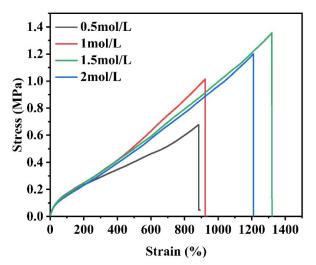


Figure S3 Mechanical properties of hydrogels with different metal ion concentrations

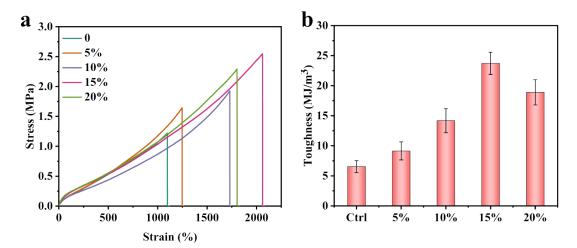


Figure S4 (a) Stress-strain curves of PAG- $Zr^{4+}$  hydrogels with different glycerol contents. (b) Toughness values of PAG- $Zr^{4+}$  hydrogels with different glycerol content.

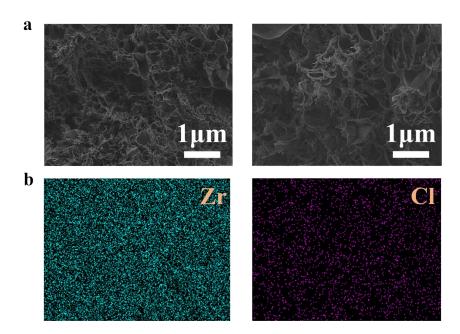


Figure S5. (a) SEM images of the hydrogel. (b) Element mapping of Zr, Cl in the  $PAEG-Zr^{4+}$  hydrogels.

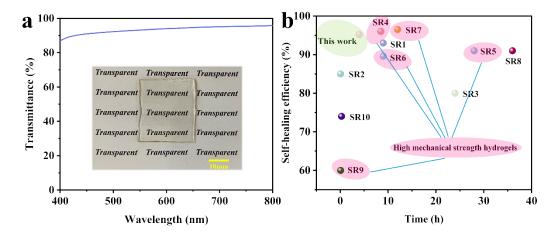


Figure S6. (a) UV-vis spectra for the hydrogel. Inset is a digital photograph of the transparent hydrogel. (b) Comparison between this work and published hydrogel in terms of self-healing time and efficiency.<sup>1-10</sup>

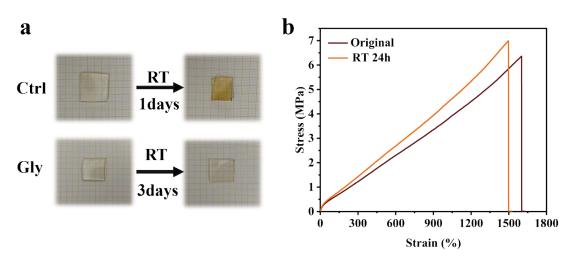


Figure S7. (a) Images of control and glycerol-based hydrogels stored at room temperature for 0 days and 3 days. (b) Stress-strain curves of  $PAEG-Zr^{4+}$  hydrogels exposed to air for 24 h at room temperature.

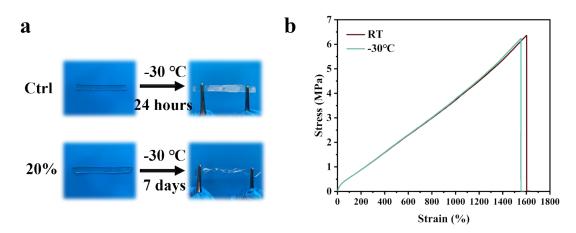


Figure S8. (a) Images of control and glycerol-based hydrogels stored at -30 °C for 24 h and 3 days. (b) Stress-strain curves of PAEG- $Zr^{4+}$  hydrogels after stored at -30 °C for 24h.

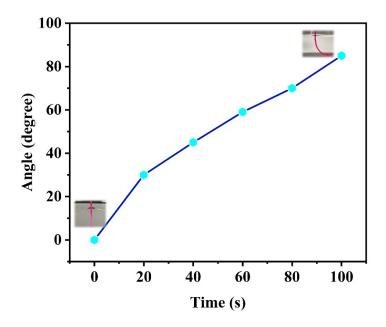


Figure S9. Bending angles of hydrogels under electrical actuation.

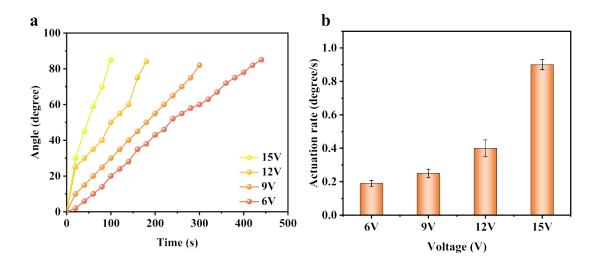


Figure S10. (a) Bending angles and (b) actuation rates of hydrogels at different voltages.

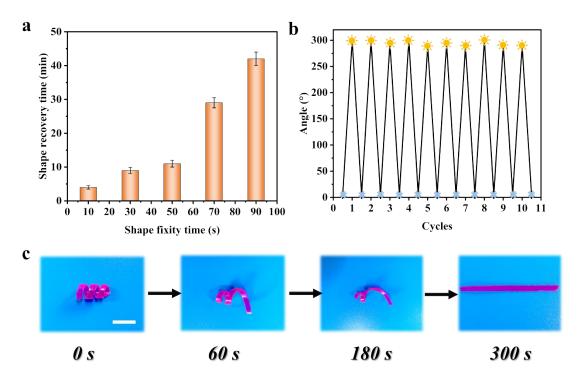


Figure S11. (a) Plot of shape recovery time and shape fixation time. (b) Cyclic memory properties of the hydrogels. (c) Recovery status of helical hydrogels at different times (scale bar = 2 cm).

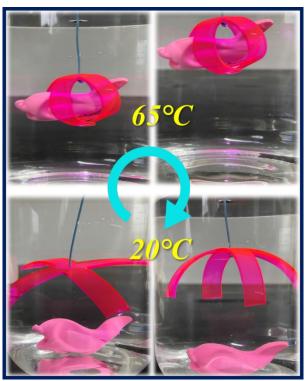


Figure S12. PAEG-Zr<sup>4+</sup> hydrogel as a catcher to catch and release model fish in different temperature environments.

#### Reference

- [1] L. Zhao, X. Li, Y. Li, X. Wang, W. Yang and J. Ren, Biomacromolecules, 2021, 22, 1273-1281.
- [2] J. Ai, K. Li, J. Li, F. Yu and J. Ma, International Journal of Biological Macromolecules, 2021, 172, 66-73.
- [3] X. Pei, H. Zhang, Y. Zhou, L. Zhou and J. Fu, Materials Horizons, 2020, 7, 1872-1882.
- [4] Y. Wang, X. Fang, S. Li, H. Pan and J. Sun, ACS Applied Materials & Interfaces, 2021, 15, 25082-25090.
- [5] T. Long, Y. Li, X. Fang and J. Sun, Advanced Functional Materials, 2018, 28.
- [6] Z. Pei, Z. Yu, M. Li, L. Bai, W. Wang, H. Chen, H. Yang, D. Wei and L. Yang, International Journal of Biological Macromolecules, 2021, 179, 324-332.
- [7] X. Fang, Y. Li, X. Li, W. Liu, X. Yu, F. Yan and J. Sun, ACS Materials Letters, 2020, 2, 764-770.
- [8] B. Zhang, X. Zhang, H. Song, D. H. Nguyen, C. Zhang and T. Liu, ACS Applied Materials & Interfaces, 2022, 14, 32551-32560.
- [9] L. Shuai, Z. H. Guo, P. Zhang, J. Wan, X. Pu and Z. L. Wang, Nano Energy, 2020,78.
- [10] M. Tavakolizadeh, A. Pourjavadi, M. Ansari, H. Tebyanian, S. J. Seyyed Tabaei,M. Atarod, N. Rabiee, M. Bagherzadeh and R. S. Varma, Green Chemistry, 2021, 23,1312-1329.