

# Supporting Information

## **Self-powered wearable sensing devices based on a flexible ammonium-ion battery with fatigue resistance and frost resistance based on a strong and tough hydrogel**

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A series of tests were carried out to quantitatively discuss the dependence of mechanical and electrical performances on the HEA, AS and Gly concentrations as well as soaking time. Fig.S2a and b show the mechanical properties of PHEA/AS hydrogels with different HEA concentrations. With the increase of HEA concentration, the crosslinked network in PHEA/AS hydrogel increased, and corresponding tensile strength and elongation showed an increasing trend. As the HEA concentration increased, the conductivity decreased (Fig.S2c). It can be explained that the increase of HEA concentration increases the crosslinked networks of PHEA/AS hydrogel, and hinders the transmission of conductive ions inside the hydrogel, resulting in the decrease in electrical conductivity of PHEA/AS hydrogel.<sup>1, 2</sup> Considered the experimental results of mechanics and electricity, HEA concentration of 60 wt% were chosen to conduct the following experiments. As shown in Fig. S3a and b that with the increase of AS concentration, both the tensile strength and elongation of the gel increased, while tensile strain showed a downward trend (Fig.S3c). The improvement of mechanical strength may be due to dehydration and high polymer content caused by the addition of AS. When the hydrogel is soaked in the AS solution, the conductive ions of  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$  enters the hydrogel, endowing the hydrogel with high conductivity. While the increase of AS concentration improves the mechanical properties, and restricts the transport of conductive ions, resulting the declines of conductivity.<sup>1, 2</sup> Therefore, based on the mechanical and electrical data, AS concentration of 2 mol/L were chosen to conduct the following tests.

Mechanical and electrical properties of PHEA/AS soaked for different time are shown in Fig. S4a-c. With the increase of soaking time, the mechanical properties of the gel were roughly improved, while the ionic conductivity of PHEA/AS hydrogel increased

and then decreased, and reached the maximum of 2.72 S/m after soaking for 12 h. PHEA/AS hydrogel with soaking time of 12 h was adopted to conduct following experiments. According to Fig. S5a and b, the tensile strength and elongation of the gel gradually increased with the increase of Gly concentration. Whereas the ionic conductivity of the gel gradually decreased with the increase of Gly content (Fig. S5c). It can be explained that the increase of Gly concentration, the interactions between PHEA and Gly increase owing to hydrogen bond formation between them, enhancing the mechanical properties of PHEA/AS hydrogel,<sup>3</sup> while the increase of crosslinked networks restricts ion transport, leading to the declines of conductivity. In brief, the optimal preparation conditions of PHEA/AS/Gly hydrogels are determined as HEA concentration of 60 wt%, AS concentration of 2 mol/L, soaking time of 12 h, and the ratio of water to Gly of 9:1.

## References

1. J. Wei and Q. Wang, *Small Methods*, 2019, **3**, 1900558.
2. R. Jin, J. Xu, L. Duan and G. Gao, *Carbohydr. Polym.*, 2021, **268**, 118240.
3. C. F. Dai, X. N. Zhang, C. Du, A. Frank, H. W. Schmidt, Q. Zheng and Z. L. Wu, *ACS Appl. Mater. Interfaces*, 2020, **12**, 53376-53384.

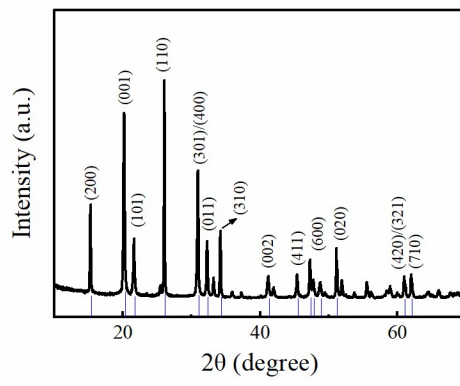


Fig. S1. XRD pattern of  $V_2O_5$ .

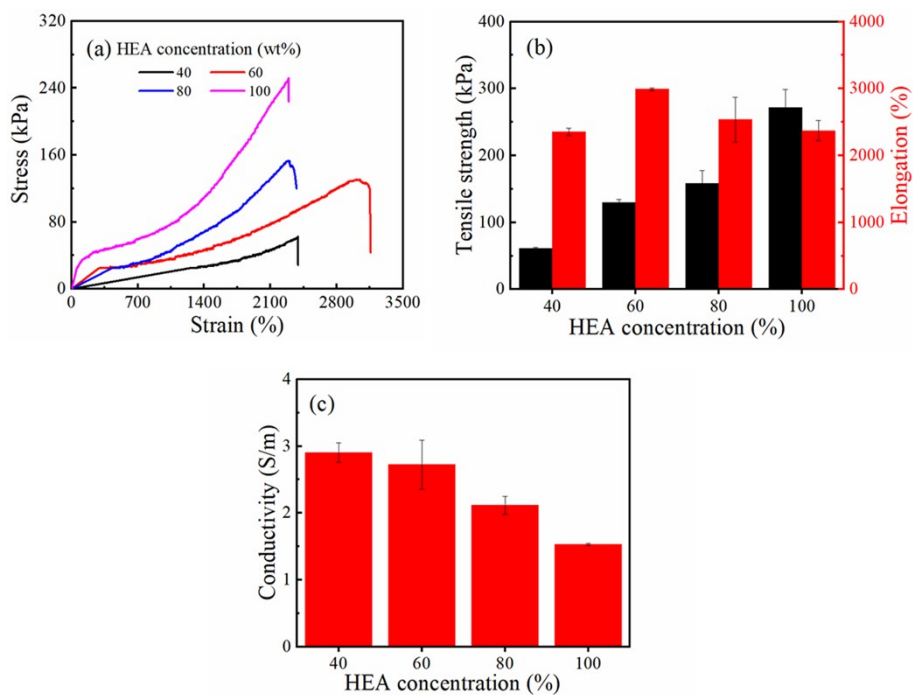


Fig. S2. (a) Tensile stress-strain curves, (b) tensile strength and elongation, (c) conductivity of PHEA/AS hydrogels with different HEA concentrations (AS concentration of 2 mol/L, soaking time of 12 h).

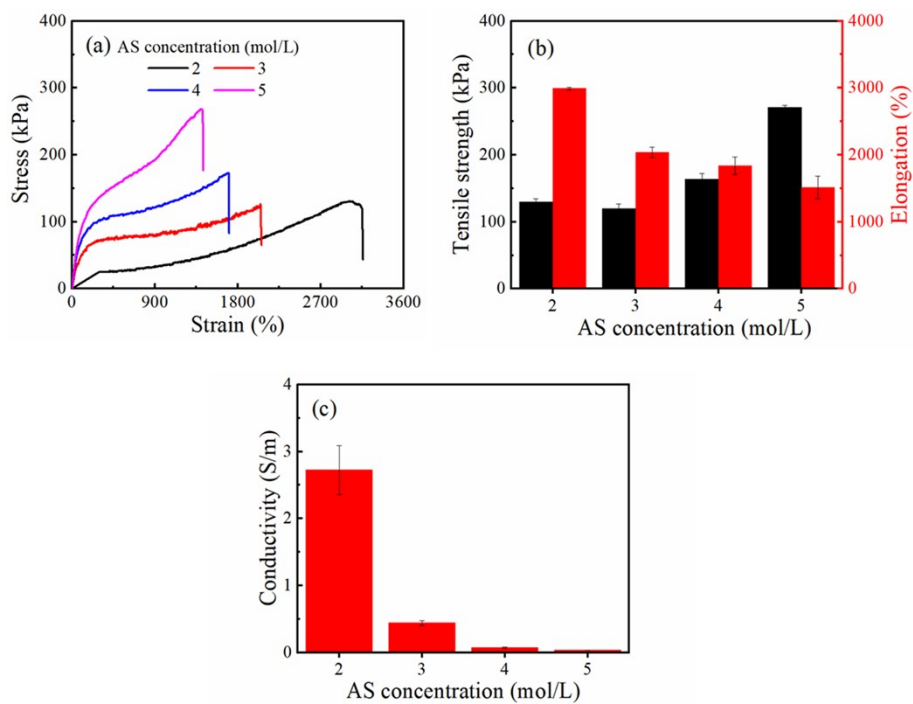


Fig. S3. (a) Tensile stress-strain curves, (b) tensile strength and elongation, (c) conductivity of PHEA/AS hydrogels with different AS concentrations (HEA concentration of 60 wt%, soaking time of 12 h).

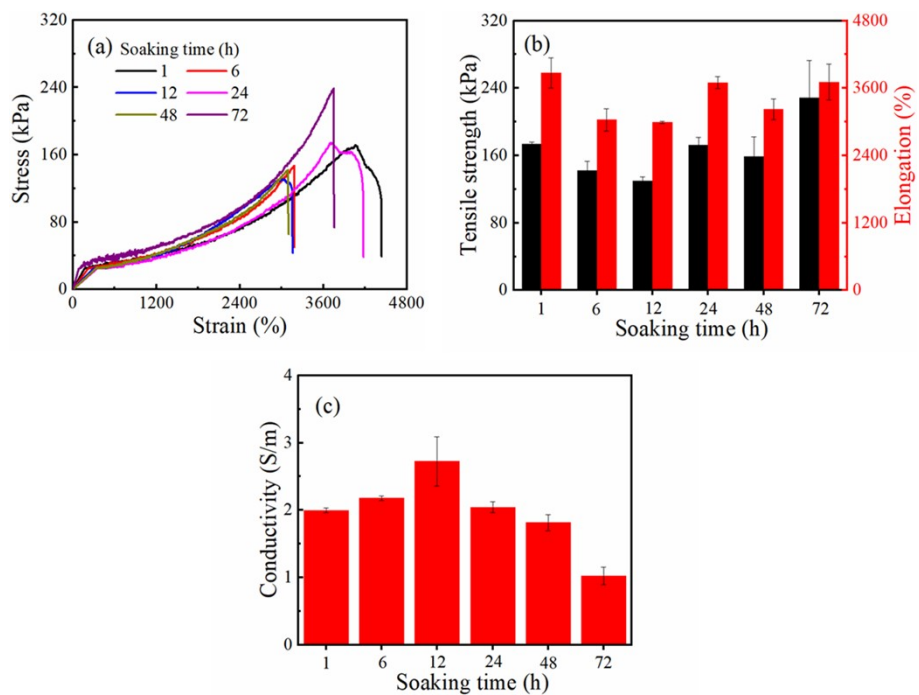


Fig. S4. (a) Tensile stress-strain curves, (b) tensile strength and elongation, (c) conductivity of PHEA/AS hydrogels with different soaking time (HEA concentration of 60 wt%, AS concentration of 2 mol/L).

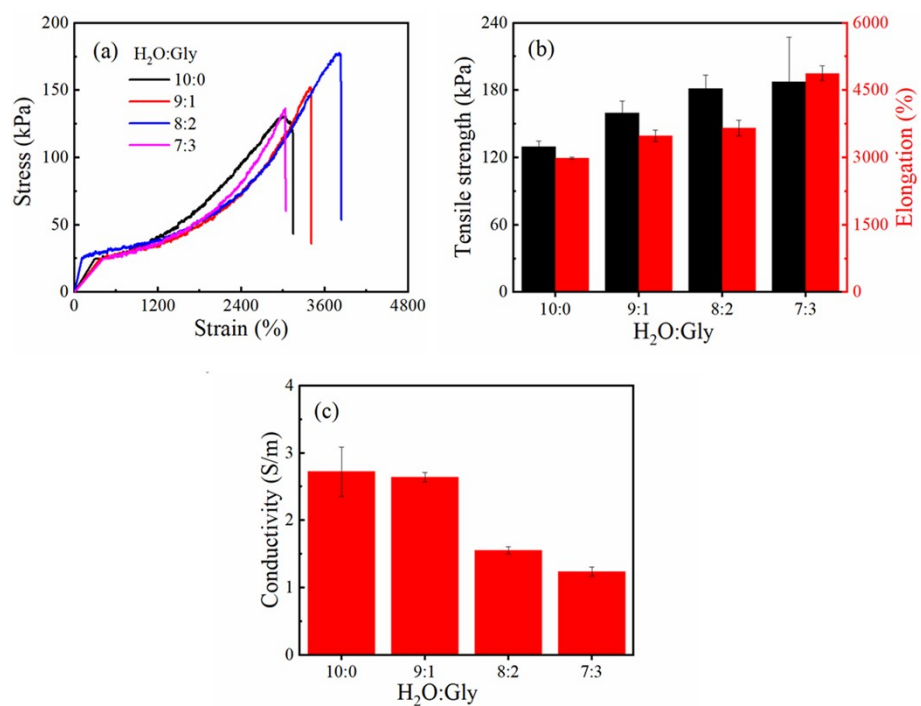


Fig. S5. (a) Tensile stress-strain curves, (b) tensile strength and elongation, (c) conductivity of PHEA/AS/Gly hydrogels with different Gly concentrations (HEA concentration of 60 wt%, AS concentration of 2 mol/L, soaking time of 12 h).



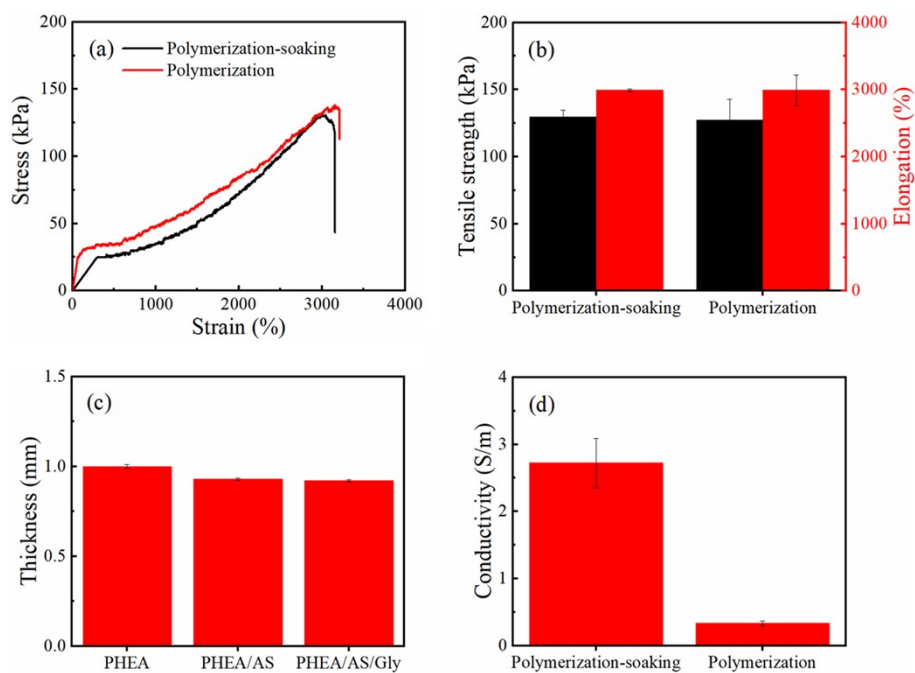


Fig. S6. (a) Tensile stress-strain curves, (b) tensile strength and elongation of PHEA/AS hydrogels with polymerization-soaking and polymerization. (c) Thickness of PHEA, PHEA/AS, PHEA/AS/Gly hydrogels with polymerization-soaking. (d) Conductivity of PHEA/AS hydrogels with polymerization-soaking and polymerization.

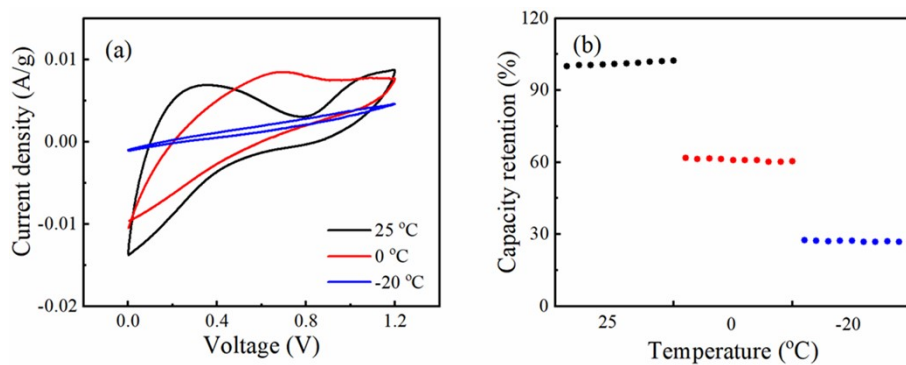


Fig. S7. (a) CV curves and (b) capacitance retention of PHEA/AS/Gly hydrogel-based flexible ammonium-ion battery at low temperatures (current density of 0.1A/g).

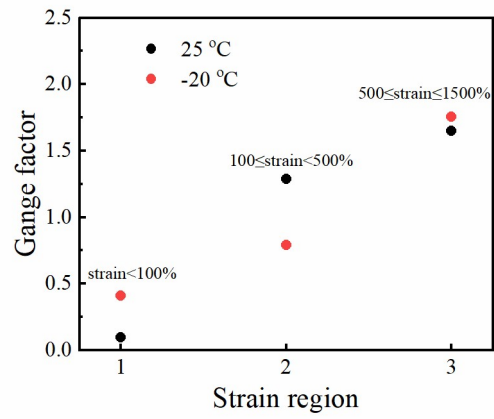


Fig. S8. Gauge factors at different tensile strains of PHEA/AS/Gly hydrogel-based flexible sensor.

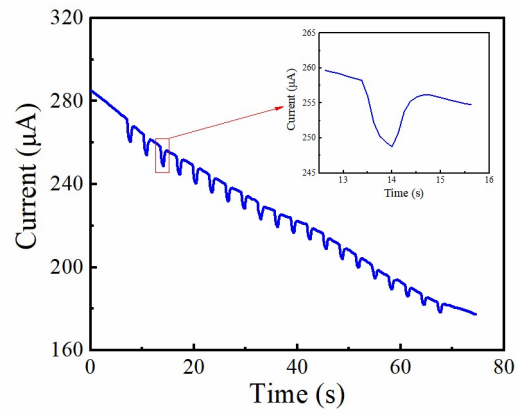


Fig. S9. Current response of self-powered sensing devices to strain of 5%.

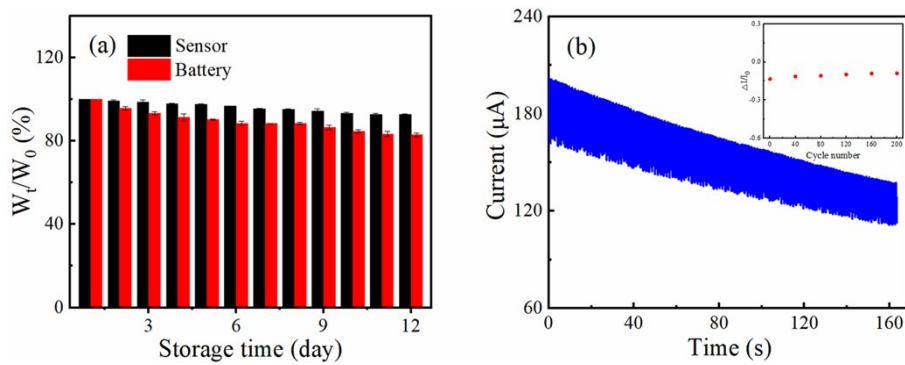


Fig. S10. (a) Weight changes of flexible ammonium-ion battery and flexible sensor placed at room temperature for several days. (b) Current response of self-powered sensing devices after storing at room temperature for 12 days to strain of 25% for 200 stretching cycles.

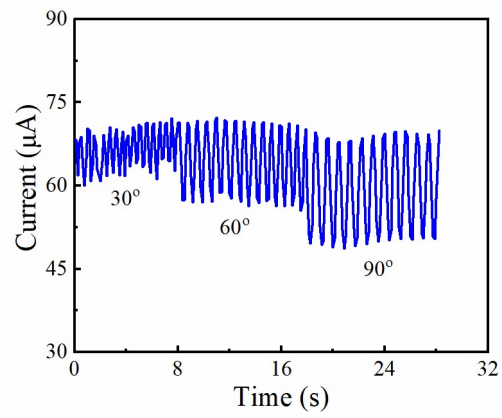


Fig. S11. Current response of self-powered sensing devices to the bending of the finger at -20 °C.