## Supporting information

## Multi-modal deformation sensing hydrogel by nerve-inspired highly

## anisotropic structure

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The <sup>1</sup>H NMR spectra of GMHA showed two new proton peaks caused by methacrylate groups at 5.22 ppm and 5.50 ppm, demonstrating the successful synthesis of GMHA.<sup>1,2</sup> The degree of methacrylatation (DM) was determined by the integration of the methyl peak and the methacrylate peak at 1.9 ppm<sup>3,4</sup>. The <sup>1</sup>H NMR spectra of GMHA with DM of 4%, 8%, and 12% was obtained.



Fig.S1 <sup>1</sup>H NMR spectra of hyaluronic acid (HA) and methacrylated hyaluronic acid (GMHA)

Fig.S2 showed the complete infrared spectrum information of the hydrogel as a supplement to Fig.2(a).



Fig.S2 FT-IR spectra of GMHA, AMH and AlgCa hydrogels.



Fig.S3 (a) SEM image of the original AlgCa hydrogel. (b) SEM image of the original GMHA hydrogel.



Fig.S4 Cross-sectional SEM image of AlgCa hydrogel with fibrous structure.

At room temperature, the prepared NS-AMH, AM<sub>4</sub>H, AM<sub>8</sub>H, and AM<sub>12</sub>H were soaked in water for 7 hours until swelling equilibrium was reached. Remove the hydrogel and wipe off excess water from the surface every hour, then weigh the mass. The swelling  $w_s - w_0$ 

rate of the hydrogel was calculated according to the formula: Swelling Ratio=  $w_0$ , where  $W_s$  and  $W_0$  were the hydrogel mass after swelling and the original hydrogel, respectively. After 7 hours of swelling test, the swelling equilibrium was reached, and the results were observed. It was found that NS-AMH had the highest swelling ratio, while the AMH's swelling rate was reduced. This indicates that AMH has better antiswelling properties.



Fig.S5 Swelling ratio curves of NS-AMH, AM<sub>4</sub>H, AM<sub>8</sub>H, and AM<sub>12</sub>H in water.

The moisturizing properties of NS-AMH and AM<sub>8</sub>H were tested. The mass change of the hydrogel over 72 hours was recorded in an open environment at room temperature. As shown in the Fig.S6, the NS-AMH hydrogel rapidly lost water within 24 hours, and the appearance shrank significantly, eventually retaining 26% of the original weight. AM<sub>8</sub>H retained 60% weight after 24 hours due to its dense fiber network that prevents the overflow of water molecules, exhibiting good moisturizing properties.



Fig.S6 Mass curves and images of NS-AMH and  $\mathsf{AM}_8\mathsf{H}$  left at room temperature for

72 hours.

The circuits consisting of  $AM_4H$ ,  $AM_8H$ , and  $AM_{12}H$  hydrogels and wires can all light light-emitting diode (LED) lamps at 4.5 V.



Fig.S7 Images of (a)  $AM_4H$ , (b)  $AM_8H$  and (c)  $AM_{12}H$  hydrogel lighting a light-emitting diode (LED) lamp at a voltage of 4.5V.



Fig.S8 The conductivities of NS-AMH, AM<sub>4</sub>H, AM<sub>8</sub>H and AM<sub>12</sub>H hydrogels.

The relative resistance changes of NS-AM<sub>8</sub>H and AlgCa hydrogel with fibrous structure were tested respectively. Under 10% strain conditions, NS-AM<sub>8</sub>H showed a certain mechanical response potential, but its ability to accurately respond was limited. In addition, NS-AM<sub>8</sub>H could not maintain the stable transmission of electrical signals, and obvious signal attenuation was observed after 3 to 5 times stretching or compression. When the same 10% strain stimulus was applied to AlgCa hydrogel with fibrous structure, the image showed that AlgCa hydrogel was able to effectively convert tensile deformation into stable electrical signal. This shows that the fiber structure does improve the hydrogel's ability to respond to tensile strain. However, the AlgCa hydrogel with fibrous structure had insufficient response to compressive strain and low sensitivity. The AMH hydrogel can respond sensitively to tensile strain and compressive strain, which shows that the pore structure further improves the strain response performance of the hydrogel. They make up for the shortcomings of AlgCa hydrogel in compressive strain response and give the hydrogel the ability to recognize three-dimensional motion signals.



Fig.S9 (a) Relative resistance change of NS-AM<sub>8</sub>H hydrogel when stretched. (b) Relative resistance changes of NS-AM<sub>8</sub>H hydrogel when compressed. (c) Relative resistance changes of AlgCa hydrogel with fibrous structure when stretched. (d) Relative resistance changes of AlgCa hydrogel with fibrous when compressed.

Hydrogels come into contact with surfaces of objects with different roughness when they are used, so it must be able to generate an electrical signal corresponding to the roughness of an external object to produce an accurate response signal. Used the AMH hydrogel to touch objects of varying roughness in everyday life, including smooth hard surfaces, sandpaper, and smooth soft surfaces. The experimental results showed that there was a significant difference in the electrical signal when touching objects with different roughness values.



Fig.S10 the response diagram of AMH hydrogel pressed on a smooth hard surface, sandpaper, and a smooth soft surface.

In addition to the macroscopic deformation related to human motion detection, the AMH can also respond to various types of micro-deformations. When writing on the hydrogel, the hydrogel can respond mechanically according to the characteristics such as writing order and strength, and convert them into different electrical signals. Electrodes were connected to the left and right sides of the prepared hydrogel, and four different letters "S, J, T, U" were written on the surface of the hydrogel. The corresponding RRC signals were significantly different (Fig. S10). Therefore, AMH hydrogel can respond to tiny mechanical stimuli and distinguish different handwritten letters.



Fig.S11 Relative resistance changes when "S, J, T, U" are written on the surface of AMH hydrogel.



Fig.S12 Sensor response diagram of the toe ball area during walking, running, and jumping for different modes of movement.

## References

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