Supplementary Information

Appendix A. Supplementary material

The detail of the materials, instruments and the preparation of reduced graphene oxide, polydopamine and different component content of the rGO/PDA-Gly-PVA composite hydrogels; the methods of material characterization, mechanical properties test, electrical properties test and calculating formula, strain sensing test.

Reduced graphene oxide reinforced PDA-Gly-PVA composite hydrogel as strain sensors for monitoring human motion

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Experimental section

Materials

Hydrochloric acid (HCl), sodium nitrate ($NaNO_3$), glutaraldehyde (GA), sodium hydroxide (NaOH), Glycerol (Gly) and graphite powder were purchased from Xilong Chemical Co., LTD. Dopamine hydrochloride (DA) was purchased from Shanghai Macklin Biochemical Co., LTD. Polyvinyl alcohol (PVA) and Tris(hydroxymethyl) aminomethane (Tris-HCl, ≥99%) were purchased from Aladdin Industrial Corporation. Sodium hydrogen sulphite ($NaHSO_4$) was purchased from Shanghai Macklin Biochemical Co., LTD. was purchased from Sichuan Xilong Science Co. LTD.

Preparation of rGO

Firstly, Graphene oxide (GO) was prepared by an improved Hummer method. Secondly, GO solution was mixed with $NaHSO_4$ under stirring in a 95 °C water bath for 5 h. After cooling down to room temperature, the lower precipitate was taken for centrifugation (8000 r/min, 2 min). The precipitate was washed with water and centrifuged several times. Finally, the black precipitate was freeze-dried to obtain rGO.

Preparation of PDA

Firstly, 1 mmol/L Tris-HCl was mixed with dopamine (DA). Meanwhile, pH value of the solution was adjusted to 8.5 with NaOH solution. After polymerization with stirring at room temperature for 24 h, the PDA solution with a concentration of 2 mg/ml was obtained.

Preparation of rGO/PDA-Gly-PVA hydrogels (PGPHs)

Firstly, rGO was put into deionized water to prepare rGO dispersion with different concentrations using sonicate for 2 h. Then, added 2 mg/ml PDA solution and glycerol. After stirring for 30 min, 0.5 ml HCl solution (1 M) and

PVA solution were added and stirred for 20 min. Next, dropped GA solution (0.25wt%) and stirred for 10 min. Finally, the solution was poured into the latex mould and then freeze-thawed to obtain rGO/PDA-Gly-PVA hydrogel. PGPHs with different contents of PVA(A), PDA(B), rGO(C), Gly(D) were designed as shown in Table S1.

Table S1 The details of samples with different contents

Experimental code	PVA / wt%	PDA / wt%	Gly / wt%	rGO / wt%
A	1.0/2.0/3.0/4.0/5.0	0.025	4	0.125
B	2.0	0.0125/0.025/0.05/1.0	4	0.125
C	2.0	0.025	4	0.05/0.1/0.25/0.5/1.0
D	2.0	0.025	0/2.5/5.0/7.5/1.0	0.125
PGPH-1	2.0	0.025	4	0.125

Material characterization

After freeze-drying of the prepared GO and rGO materials, the products were analysed by X-ray diffractometer (XRD, D8-ADVANCE, Brucker), and the degree of reduction was analysed by the characteristic peaks generated. Fourier transform infrared spectroscopy (FTIR, Nicolet IS10, Thermo Fisher) was used to characterize the synthesized hydrogel materials, and the absorption spectra of the molecules were analysed to obtain the functional groups and chemical bonding in the hydrogels. The Raman spectroscopy (LabRAM, HR,Horiba) of the sample materials were analysed and the characteristic structures of the materials could be analysed. The surface morphology of the material in the sample preparation was recorded using field emission scanning electron microscopy (SEM, S-4800, Hitachi).

Mechanical properties test

The hydrogel precursor solution was poured into a special barbell-shaped mould to prepare a sample with relatively the same morphology. The specific details of the mould are shown in Figure S1, and optical images of the sample and the mould are shown in Figure S2. The stress-strain of the hydrogel and the maximum tensile length was measured through tensile tests using the universal tensile machine (AG-X 10KN, SHIMADZU).

Strain rate indicates the rate of sample undergoes deformation; the value is the testing speed divided by the sample gauge length (L_0):

$$\text{Strain rate} = \text{Testing speed}/L_0 \tag{1}$$

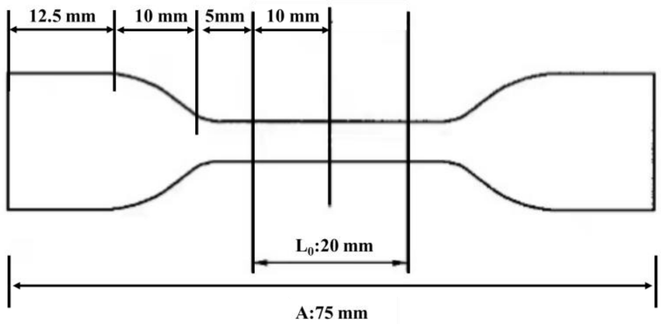

Figure S1. The details of barbell-shaped sample

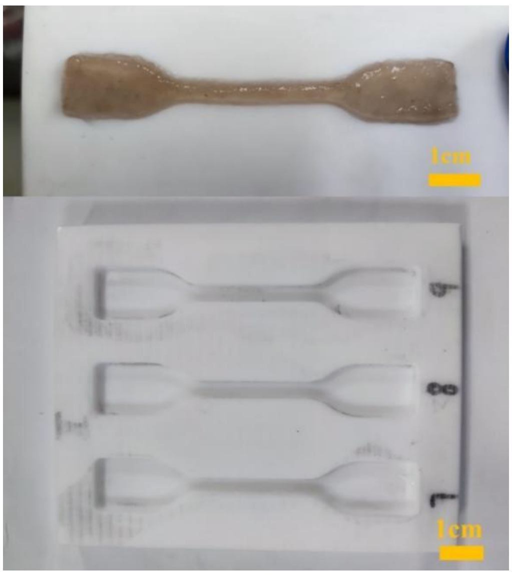

Figure S2. The barbell-shaped sample and the mould

Electrical properties test

(1) The hydrogel precursor solution was poured into a mould of size 20mm×40mm×10mm and prepared into a rectangular block with relatively the same shape, and the impedance test of the hydrogel was carried out by using an electrochemical workstation (Vertex.One.EIS, Ivium), and the conductivity of the hydrogel was calculated according to the following equation (2)

$$\sigma = 1/\rho = L/RS \tag{2}$$

Where R is the resistance of the hydrogel (Ω); ρ is the resistivity of the hydrogel(Ω·m); L is the length of the hydrogel sample (m); S is the area of the hydrogel sample in contact with the electrode (m^2); σ is the conductivity of the hydrogel sample (S/m).

(2) The hydrogel precursor solution was poured into a special barbell-shaped mould to prepare a sample with relatively the same morphology, and the relative resistance change rate ($\Delta R/R_0$) of the hydrogel sample was measured under different stretching ratios, which was calculated as Equation (3), while the gauge factor (GF) of the hydrogel sample was calculated as Equation (4).

$$\Delta R/R_0 = \frac{|R_0 - R|}{R_0} \tag{3}$$

$$GF = \frac{\delta(\Delta R/R_0)}{\delta\varepsilon} \tag{4}$$

Where R_0 is the resistance value of the hydrogel in the initial state (Ω); R is the resistance value of the hydrogel in the stretched loaded state (Ω); ε is the deformation factor of the hydrogel when it is stretched, no unit of measurement.

Strain sensor detection test

The hydrogel is prepared into a block with silver wire electrode to form a wearable sensor that performance good adhesion. The sensor is attached to a person's joints and throat, and detects responses to human actions, including human limb behavior and small vocal cord vibrations.

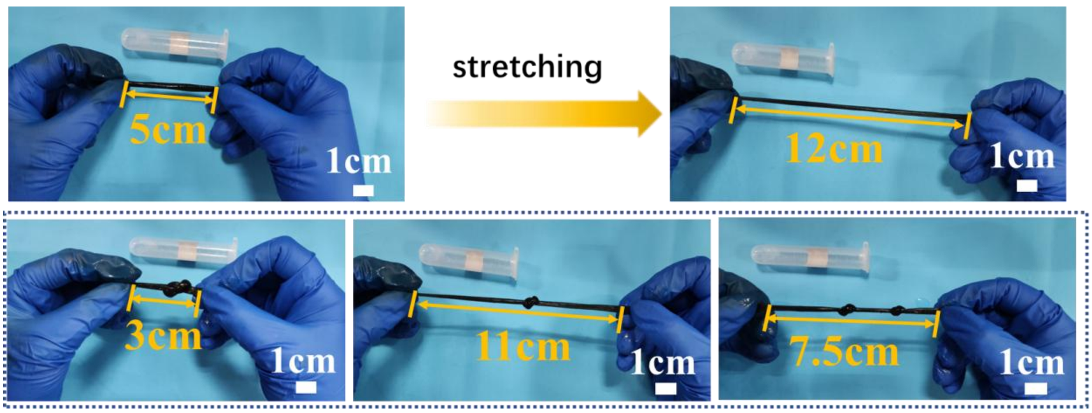

Figure S3. Display of tensile properties of hydrogels

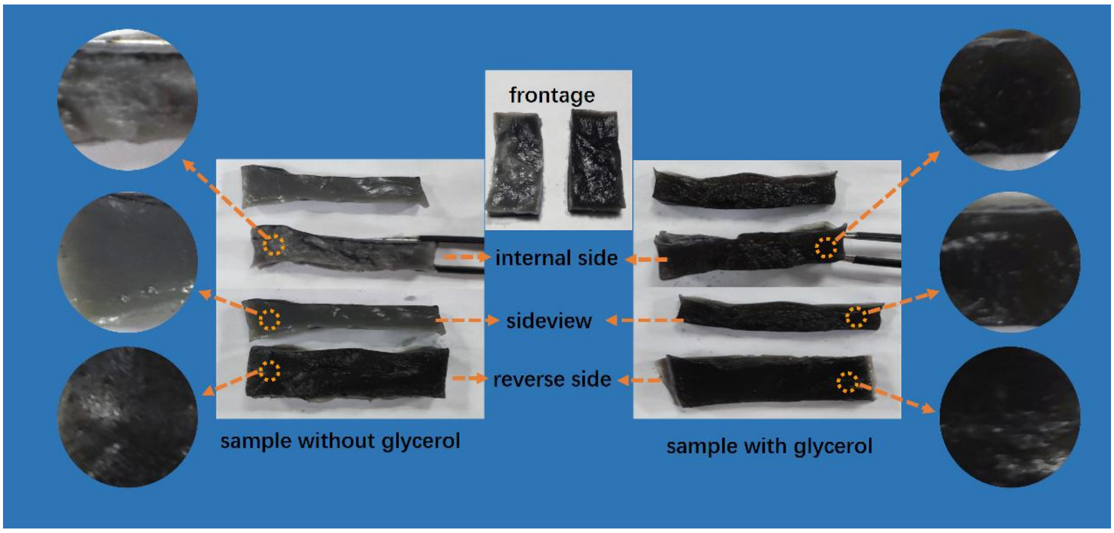

Figure S4. Comparison of different parts of samples with and without glycerol

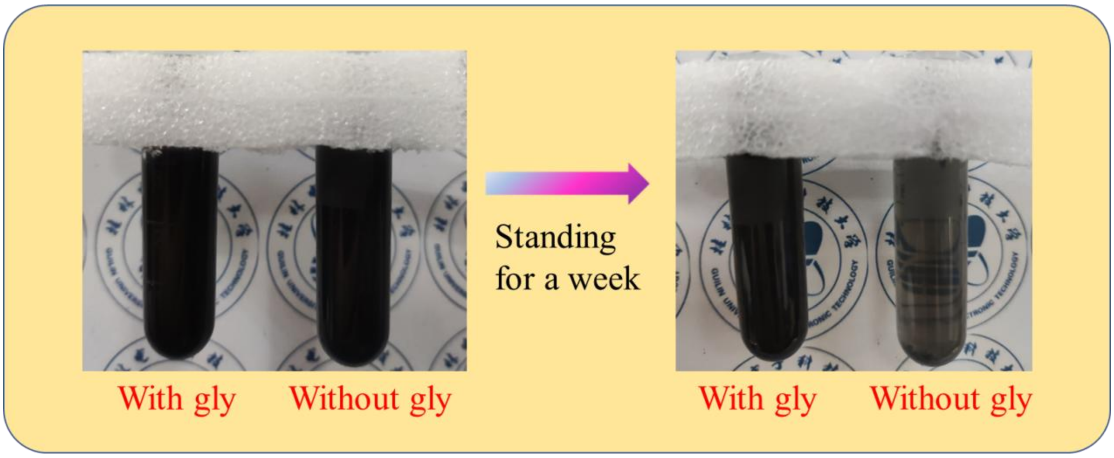

Figure S5. Comparison of solution with and without glycerol after standing for a week

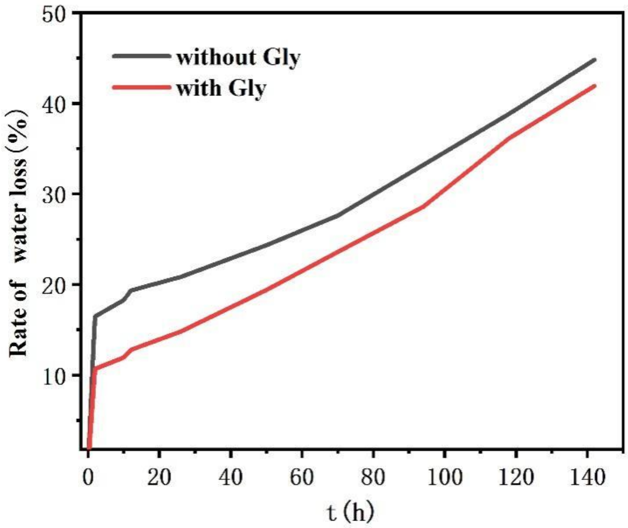

Figure S6. Water loss rate curve of the sample with and without glycerol

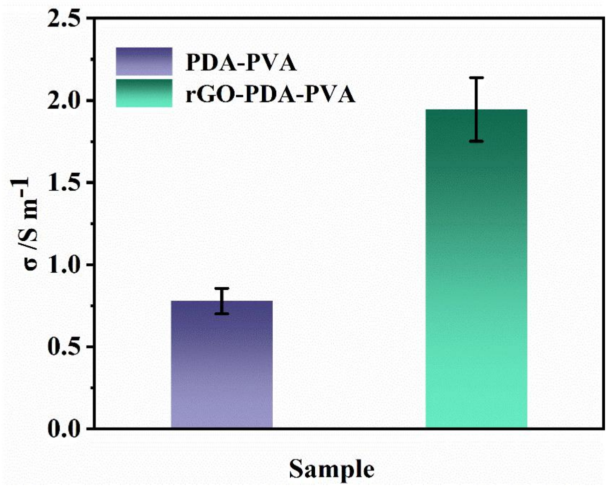

Figure S7. The conductivity of hydrogels with and without rGO.