

Supporting Information

Extremely Stretchable, Stable and Durable Strain Sensors Based on Double-Network Organogels

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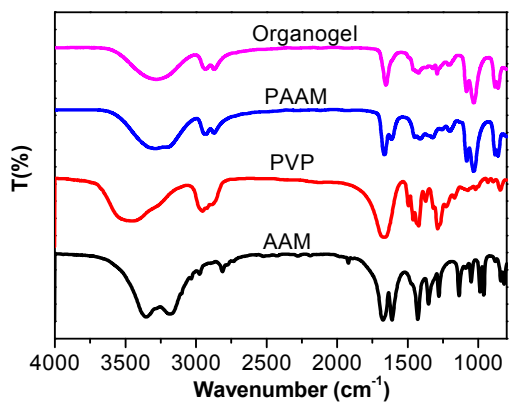


Figure S1. FT-IR spectra of AAM monomer, PVP, PAAM and organogel.

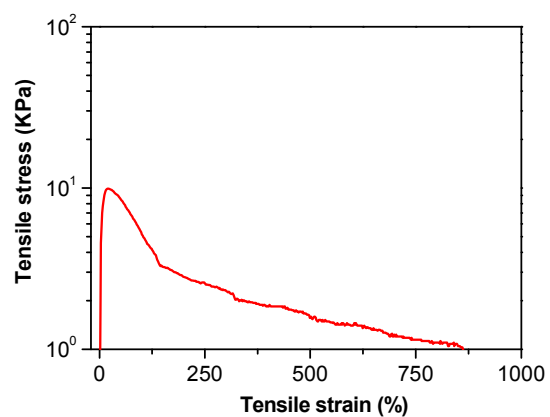


Figure S2. Tensile strain-stress curve of PAAM/PVP/EG organogel with a crosslinker weight ratio of 0.13 wt% (PVP, 2.8 wt%).



Figure S3. Optical images of as-prepared extremely stretchable organogel (a) before and (b) after stretching with the original length of 5 mm.

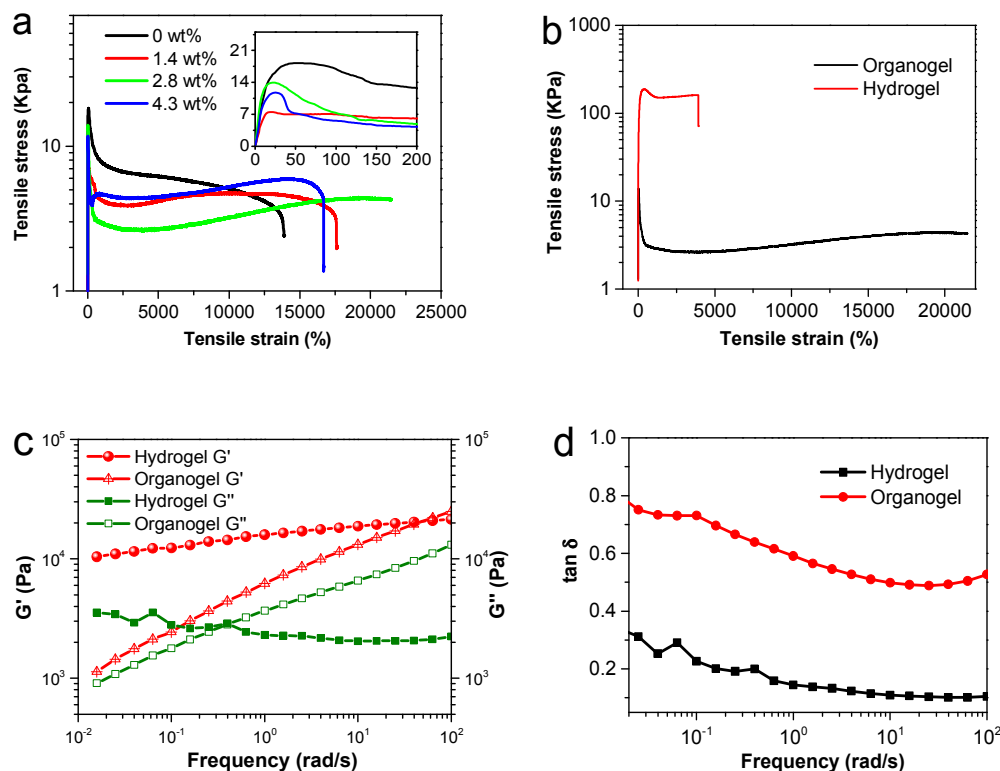


Figure S4. (a) Tensile strain-stress curves of PAAM/PVP/EG organogels prepared with various PVP concentrations (PEGDA400 0.25 wt%). When PVP was 2.8 wt%, the organogel exhibited the highest stretchability. Unless otherwise stated, the organogels used in this work were prepared with PVP concentration of 2.8 wt%. (b) Comparison of tensile stress-strain curves of organogel and hydrogel (PEGDA400 0.25 wt%, PVP 2.8 wt%). (c) Dynamic mechanical behaviors of organogel and hydrogel. (d) The loss factor ($\tan \delta = G''/G'$) of hydrogel and organogel.

Supplementary Nots: Further proof the crosslinking density of organogel was lower than that of hydrogel.

As-prepared hydrogel and organogel has the same components except for liquid constituent. Hence, the significant differences of stress-strain curve are caused mainly by noncovalent interaction. To evaluate their network structures based on noncovalent interactions, we approximately estimated their crosslinking densities by the theory of rubber elasticity:

$$\sigma = NRT(\lambda - \frac{1}{\lambda^2}) \quad (1)$$

where σ is the force per unit unstrained cross-sectional area, N represents the effective crosslinking density, R is the gas constant, T is absolute temperature, and λ is the elastic extension ratio. According to stress-strain curves of the organogel and hydrogel, the elastic elongation ratio λ is 1.18 (at elastic strain of 59%), T is 298 K, R is 6.314. Correspondingly, stresses are 17.79 and 41.35 KPa for organogel and hydrogel, respectively. The calculated crosslinking densities N are listed in Table S1. Apparently, the crosslinking density of organogel was lower than that of hydrogel, which reduced the rigidity of network and increased its elasticity.

Table S1. Approximately estimated crosslinking density of the organogel and hydrogel.

	Stress (KPa)	λ	N (mol/m ³)
Organogel	13.79	1.18	12.25
Hydrogel	41.35	1.18	36.72

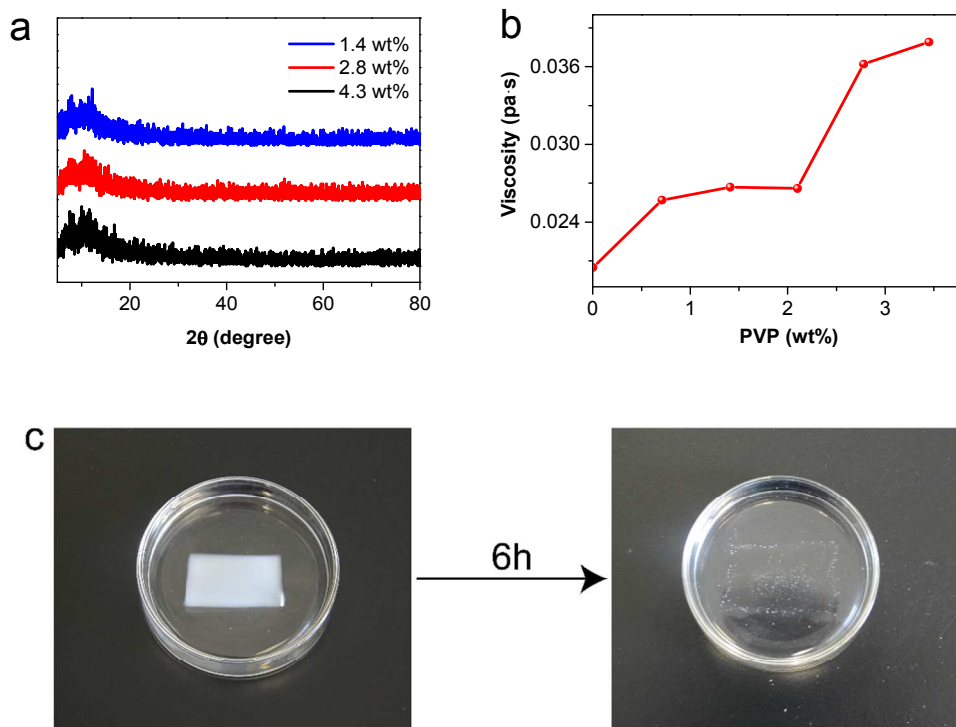


Figure S5. (a) XRD patterns of PAAM/PVP/EG organogels prepared with different PVP wt%. (b) Viscosities of organogel precursor as a function of PVP concentration. (c) After putting organogel into deionized water for 6 h, the opaque organogel became transparent, indicating disentanglement of polymer chains.

Supplementary Nots: polymer (PVP and PAAM) concentration far exceeded the critical entanglement concentration

To discuss the formation of polymer chain-entanglement in PAAM/PVP/EG organogels, we measured the overlapping concentration C^* of polymer chains in precursor solution where the viscosity increased dramatically. Clearly, the viscosity of precursor enhanced suddenly when PVP concentration increased from 2.1 wt% to 2.8 wt%, as shown in Figure S9. Because the organogels used in this work were prepared with PVP concentration of 2.8 wt%, it is indicated

that the polymer chains concentration of precursor solution reached to the overlapping concentration C^* . The corresponding repeat unit is ~ 0.29 mol/L.

After polymerization, a large amount of PAAM chains was formed. As AAM monomer concentration was 27.78 wt% in the present work, which corresponded to the repeat unit of 4.52 mol/L. This value is 15 times higher than the overlapping concentration C^* . Commonly, the critical entanglement concentration C_e is about 4-5 times of the overlapping concentration. Therefore, it is implied that polymer (PVP and PAAM) concentration far exceeded the critical entanglement concentration and polymer chain-entanglement occurred.

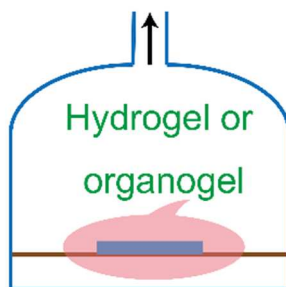


Figure S6. Schematic illustration of the experimental setup for measuring weight change of hydrogel and organogel in the vacuum chamber (0.09 atm).

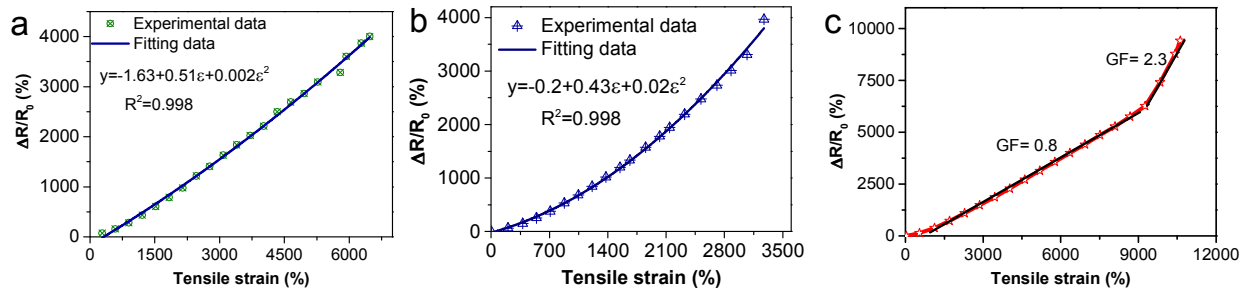


Figure S7. (a) Regression equation of the relative resistance versus strain for the sensor prepared with 0.38 wt% PEGDA400. (b) Regression equation of the relative resistance versus strain for the sensor prepared with 0.50 wt% PEGDA400. (c) Gauge factors of the strain sensor prepared with 0.25 wt% PEGDA400.

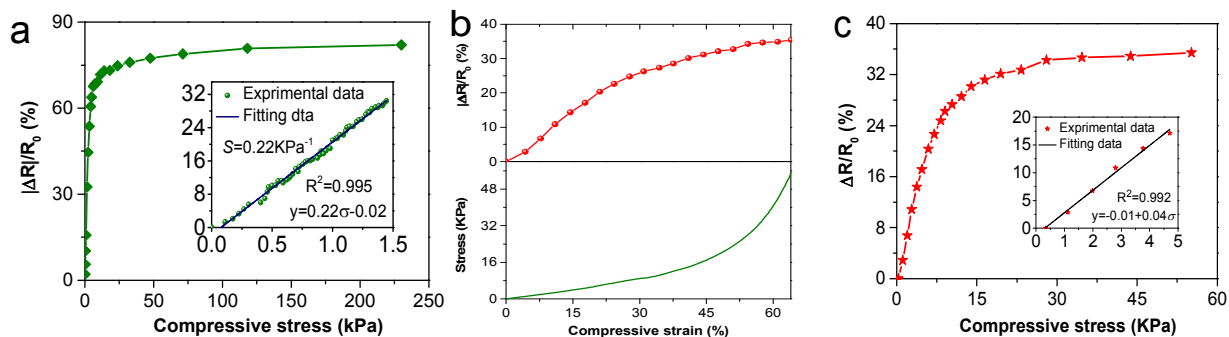


Figure S8. (a) The corresponding dependence of resistance change on applied compressive stress. The inset shows the fitted curve within 1.5 kPa stress. The inset shows the fitted curve below 1.5 kPa stress, revealing a pressure sensitivity of 0.22 kPa⁻¹. (b) Compressive stress-strain and relative resistance-strain curves of the sensor prepared with 0.5 wt% PEGDA400. (c) The compressive stress-dependent resistance change the sensor prepared with 0.5 wt% PEGDA400. The inset shows the fitted curve below 5 kPa stress, revealing a pressure sensitivity of 0.04 kPa⁻¹.

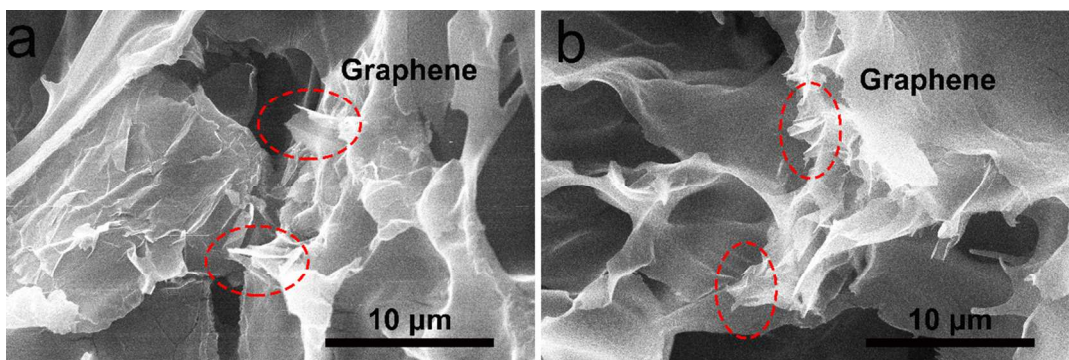


Figure S9. (a)(b) Cross-sectional SEM image of the strain sensor.

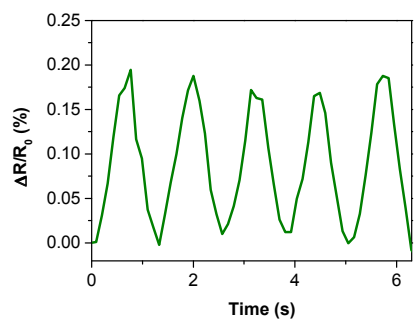


Figure S10. The relative resistance variation of strain sensor by applied small tensile strain between 0 and 0.5%, loading rate of 50 mm min^{-1} were used for measurement.

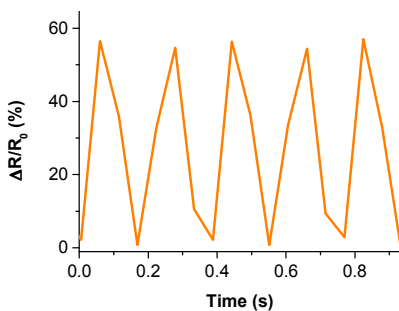


Figure S11. The relative resistance variation of strain sensor by applied tensile strain between 0 and 80% at a frequency of 3 Hz for the sensor with 0.50 wt% PEGDA400.

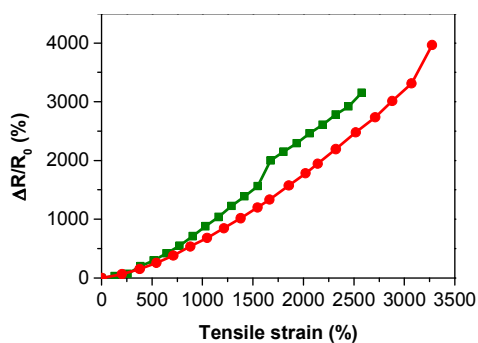


Figure S12. The relative resistance-strain curves of strain sensors prepared with different batches (0.50 wt% PEGDA400).

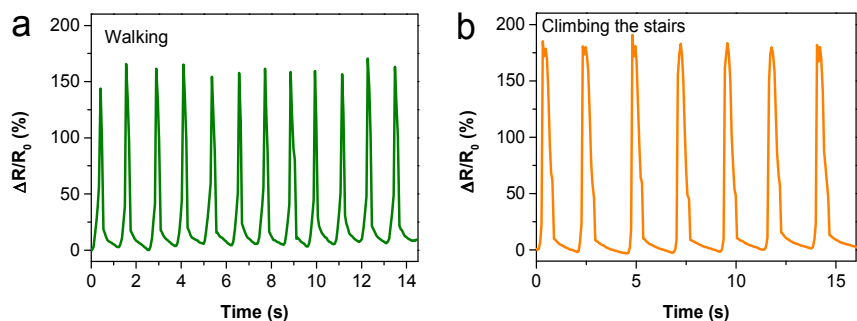


Figure S13. Relative resistance changes of the strain sensors for detecting knee joint movements during walking (a) and climbing the stairs (b).