Supporting Information

Freezing-tolerant, Widely Detectable and Ultra-sensitive Composite Organohydrogel for Multiple Sensing

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Table S1 Preparation of different hydrogels.

Sample	Soaking solution		Soaking time (d)
	$CaCl_2$ (wt/v)	H_2O/glycerol (v/v)	
Pre-hydrogel (a–No Soaking)	-	-	-
Organohydrogel–1 (b–1:1)	0.3	1:1	3
Organohydrogel–2 (c–2:1)	0.3	2:1	3
Hydrogel (d–1:0)	0.3	1:0	3

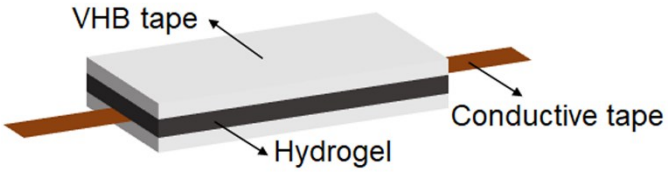

Figure S1. Schematic illustration of the sandwiched hydrogel strain sensor structure.

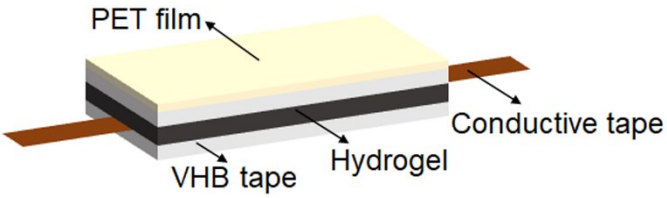

Figure S2. Schematic illustration of the sandwiched hydrogel pressure sensor structure.

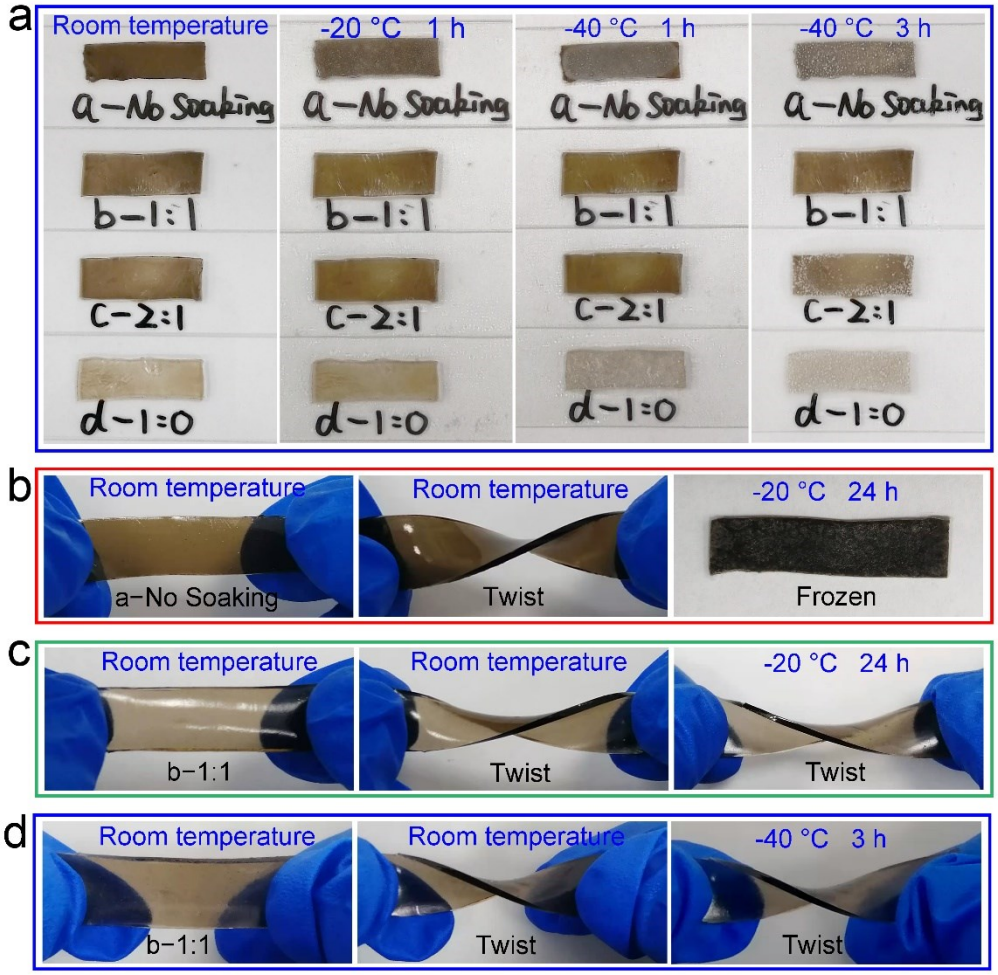

Figure S3. (a) Digital image of four different samples by placing them under different environments: room temperature, -20 °C for one hour, -40 °C for one hour, -40 °C for three hours. (b) Photographs of hydrogels (a−No Soaking) twisted at room temperature and frozen after storage for 24 h at -20°C. (c) Photographs of hydrogels (b−1:1) are twisted at room temperature and after storage for 24 h at -20 °C. (d) Photographs of hydrogels (b−1:1) is twisted at room temperature and after storage for 3 h at -40°C.

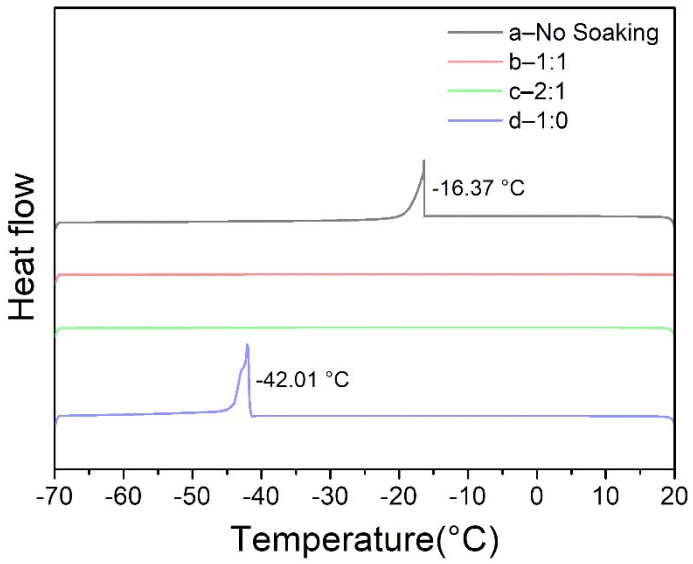

Figure S4. DSC results of hydrogels.

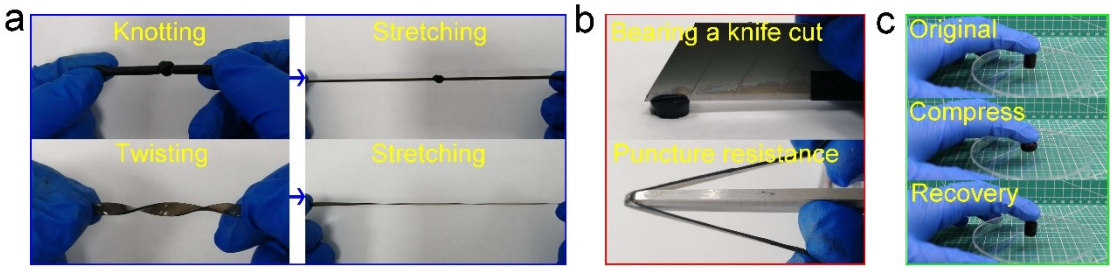

Figure S5. Mechanical properties of the PDA–rGO/SA/PAM composite hydrogel. Photos of hydrogel: (a) stretching after knotting and twisting, (b) bearing the pressure of a knife and a blunt-edged scissor, and (c) compressing.

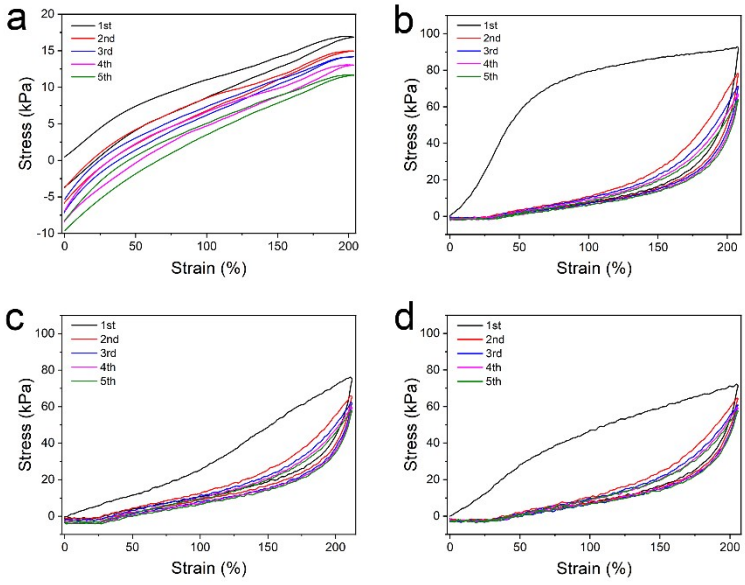

Figure S6. Strain-stress cyclic curves of (a) a–No Soaking, (b) b–1:1, (c) c–2:1 and (d) d–1:0 hydrogels.

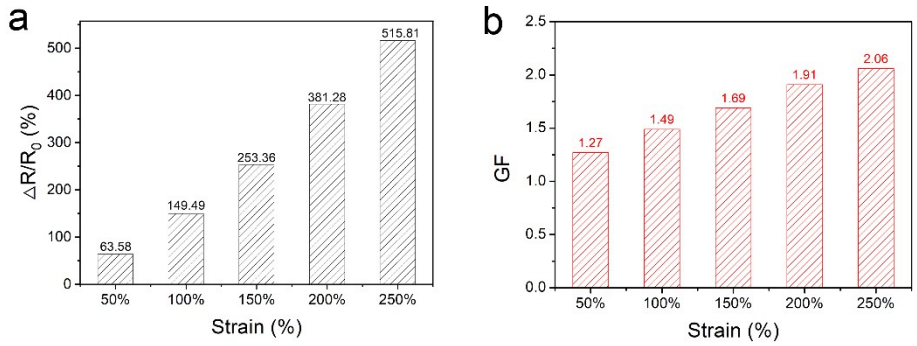

Figure S7. The real-time resistance variation (a) and sensitivity (b) at different strains (50%, 100%, 150%, 200%, 250%).

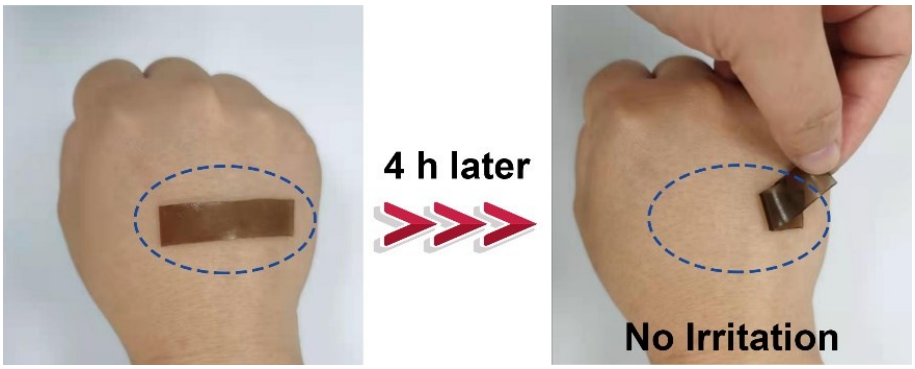

Figure S8. No irritation on human skin was detected after attaching the hydrogel for 4h indicating the hydrogel is safe to human skin.

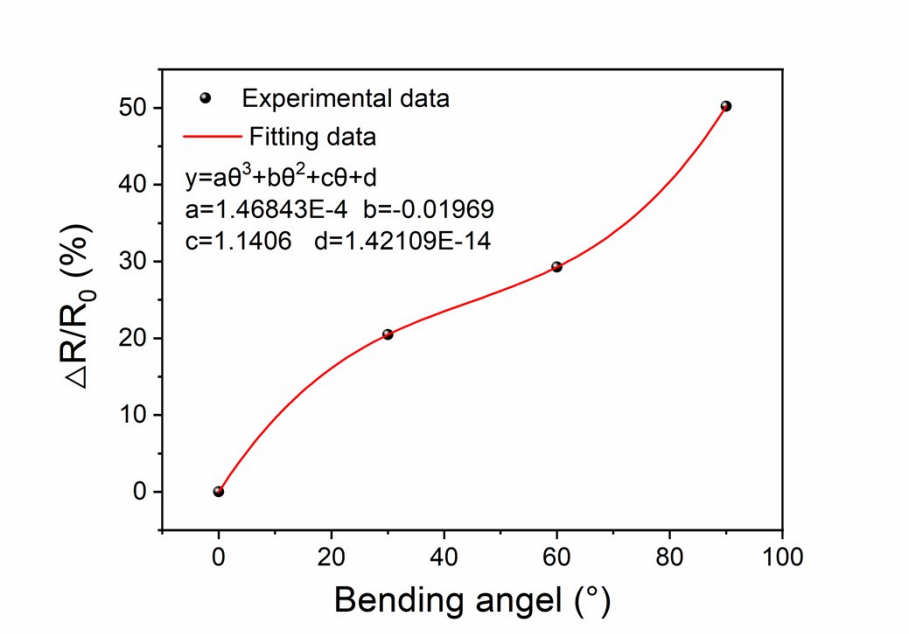

Figure S9. The fitting curve and cubic function relationship between the relative resistance change ($\Delta R/R_0$) and bending angles.

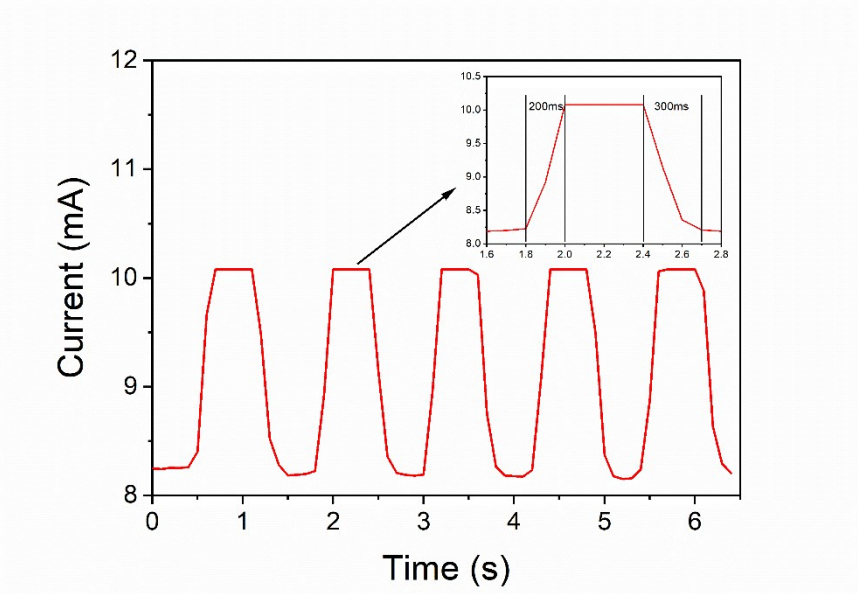

Figure S10. Response and release behavior of the strain sensor as the index finger bend.

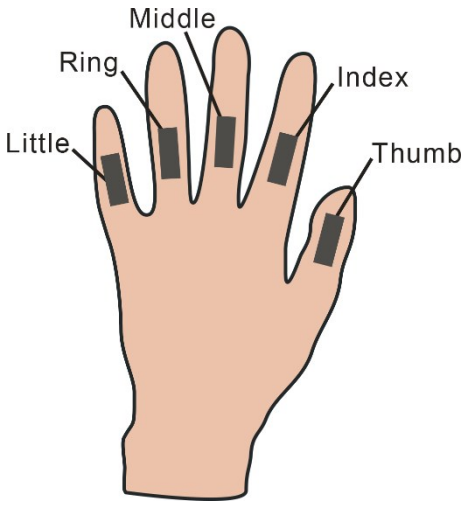

Figure S11. Schematic for a situation of five sensors (thumb, index, middle, ring, little).

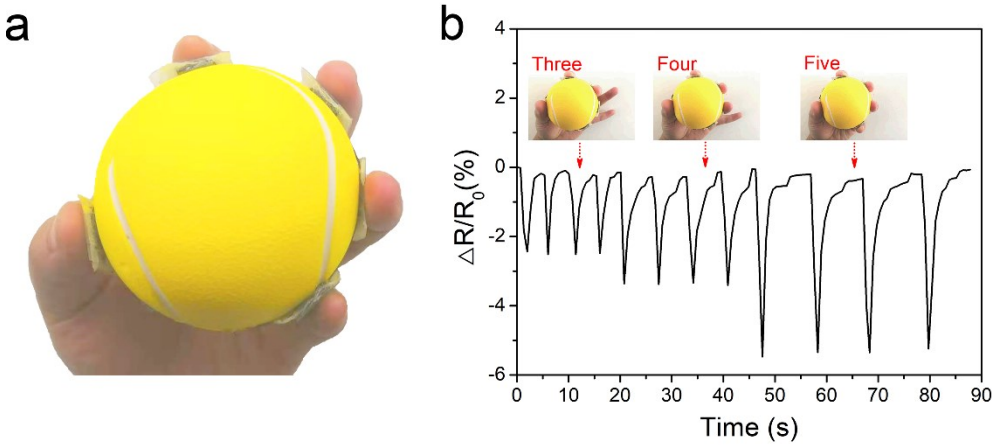

Figure S12. (a) Schematic for a situation of five sensors on the ball. (b) The real-time resistance changes corresponding to three, four, and five fingers pressing the ball.

Table S2. Comparison in the properties of hydrogel-based sensors based on different materials.

Flexible sensor composition	Tensile strength (kPa)	Sensitivity (strain, GF)	Temperature sensing range (°C)	Anti-freezing properties	Ref.
PDA–rGO/SA/PAM organohydrogel–1	143.2	0-250%, 2.09	-20 ~ 60	-20 °C, 24 h; -40 °C, 3 h	This work
PANI NFs/ PAA/Fe^{3+}	35.68	0-150%, 1.16	40 ~ 110	No	ACS Nano, 2020 [1]
PVA/Gly/CB/CNT	4800	0-700%, 2.01	30 ~ 80	-20 °C, 24 h	ACS Appl Mater Interfaces, 2020 [2]
PAM/carrageenan Gly-organohydrogels	36	Not given	25~ 102	-18 °C, 24 h	ACS Appl Mater Interfaces, 2020 [3]
PAAm/SA/CNT/$CaCl_2$	271.68 ± 6.04	0-400%, 3.125	No	-20 °C	ACS Appl Mater Interfaces, 2020 [4]
Gelatin/PAAm-oxCNTs	710	0-250%, 1.50	No	No	Chemical Engineering Journal, 2020 [5]
PAA/CS/GO/Gly	226.2 ± 30.05	0-80%, 1.138	No	-20 °C	Journal of Materials Chemistry C, 2019 [6]

References

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