Supplementary Information

A polyacrylamide/gelatin/tannic acid-modified carbon nanotubes double network hydrogel with skin temperature-triggered adhesion and high sensitivity for wearable sensors

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Table S1. Recipe for hydrogel samples

Hydrogel	Gelatin g	H_2O ml	TA-CNTs ml	CNTs ml	AM g	MBA g	APS g
PAM	0	40	0	0	8	0.016	0.08
PAM/Gelatin$_1$	0.4	40	0	0	8	0.016	0.08
PAM/Gelatin$_2$	0.8	40	0	0	8	0.016	0.08
PAM/Gelatin$_3$	1.2	40	0	0	8	0.016	0.08
PAM/Gelatin$_4$	1.6	40	0	0	8	0.016	0.08
PAM/Gelatin$_5$	2	40	0	0	8	0.016	0.08
PAM/Gelatin$_4$/TA-CNTs$_{0.06}$	1.6	36	4	0	8	0.016	0.08
PAM/Gelatin$_4$/TA-CNTs$_{0.09}$	1.6	34	6	0	8	0.016	0.08
PAM/Gelatin$_4$/TA-CNTs$_{0.12}$	1.6	32	8	0	8	0.016	0.08
PAM/Gelatin$_4$/TA-CNTs$_{0.15}$	1.6	30	10	0	8	0.016	0.08
PAM/Gelatin$_4$/CNTs$_{0.12}$	1.6	32	0	8	8	0.016	0.08

Fig. S1 Dispersibility of CNTs and TA-CNTs in water.

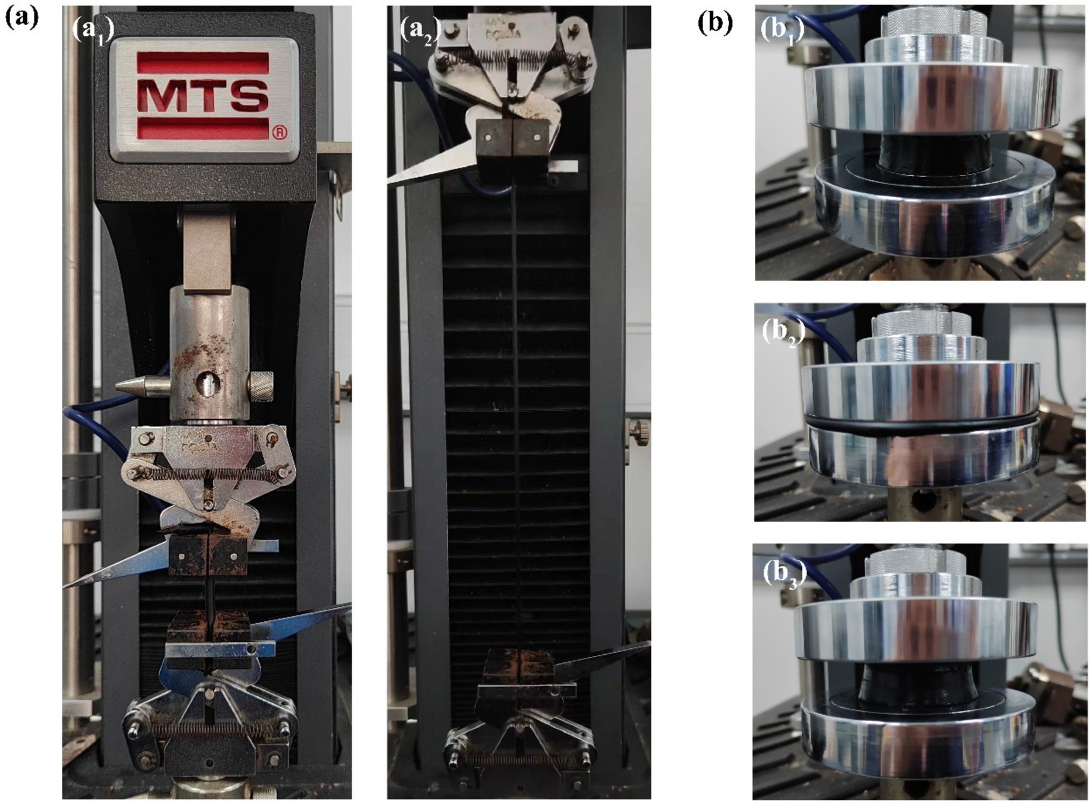

Fig. S2 (a) Images of the PAM/Gelatin$_4$/TA-CNTs$_{0.12}$ hydrogel tensile test: (a$_1$) before, (a$_2$) after; (b) Images of the PAM/Gelatin$_4$/TA-CNTs$_{0.12}$ hydrogel compression test: (b$_1$) before, (b$_2$) ε=80%, (b$_3$) after.

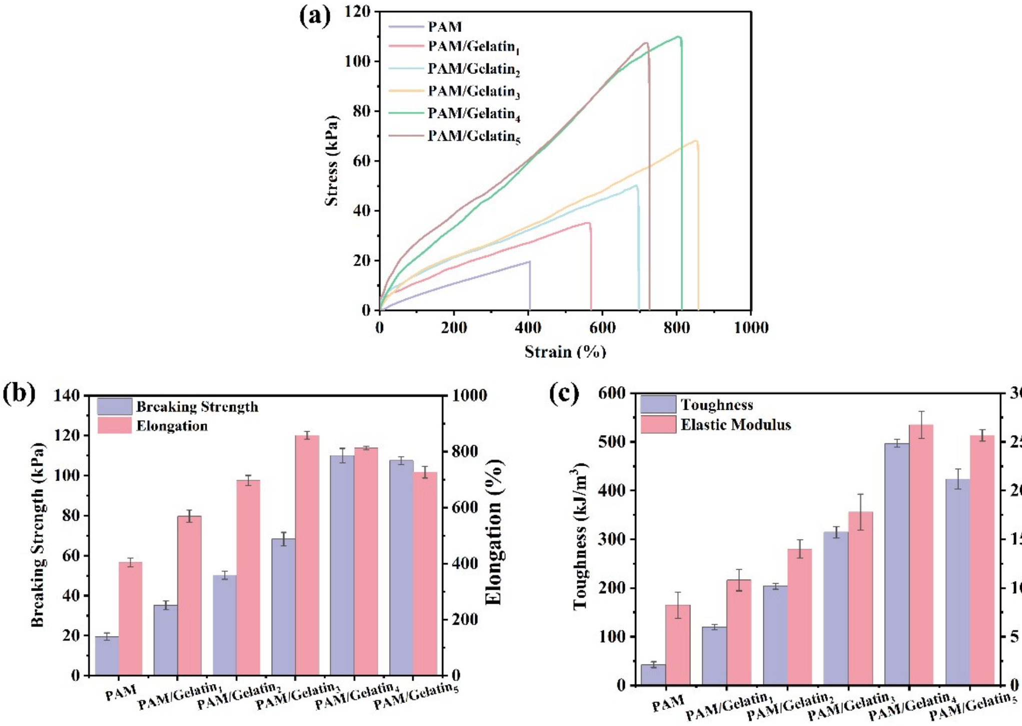

Fig. S3 (a-b) Tensile stress-strain curves of the PAM and PAM/Gelatin hydrogels (tested with Gelatin as the variable) and (c) corresponding toughness and elastic modulus.

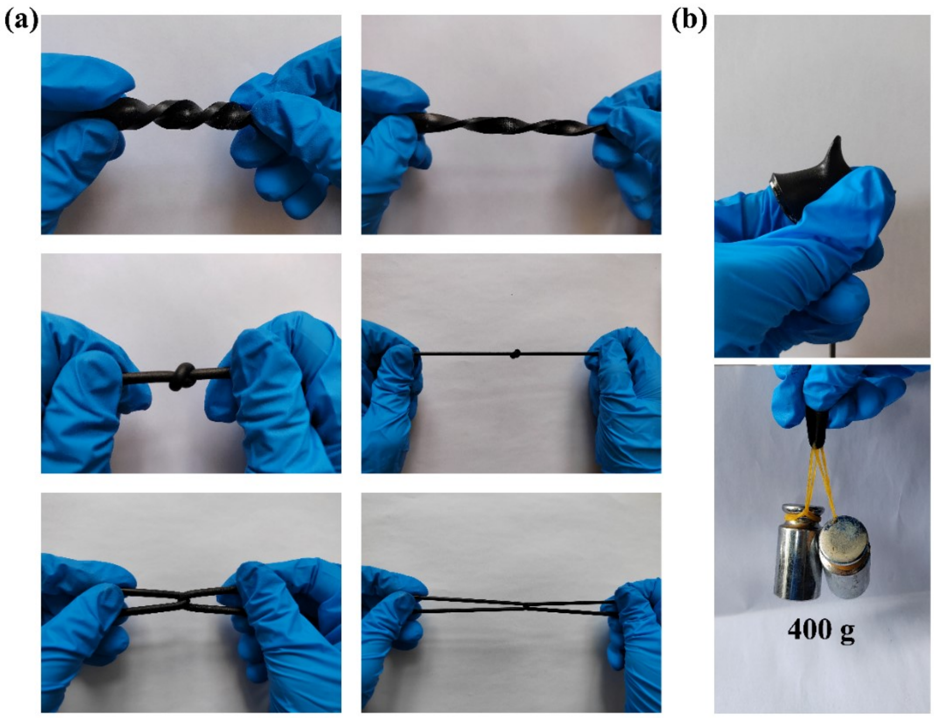

Fig. S4 (a) Curling, knotting and cross-stretching of PAM/Gelatin/TA-CNTs hydrogel; (b) PAM/Gelatin/TA-CNTs hydrogel resists puncture and withstands 400g weights without breaking.

Swelling tests

The hydrogel sample was dried to constant weight, and the initial mass W_0 was recorded. Then the sample was swollen in pure water and taken out at the same time interval. The surface moisture was absorbed by filter paper and weighed as mass Wt. The swelling ratio of the hydrogel was calculated according to equation (1):

$$Swelling\ ratio = \frac{W_t - W_0}{W_0} \times 100\% \#(1)$$

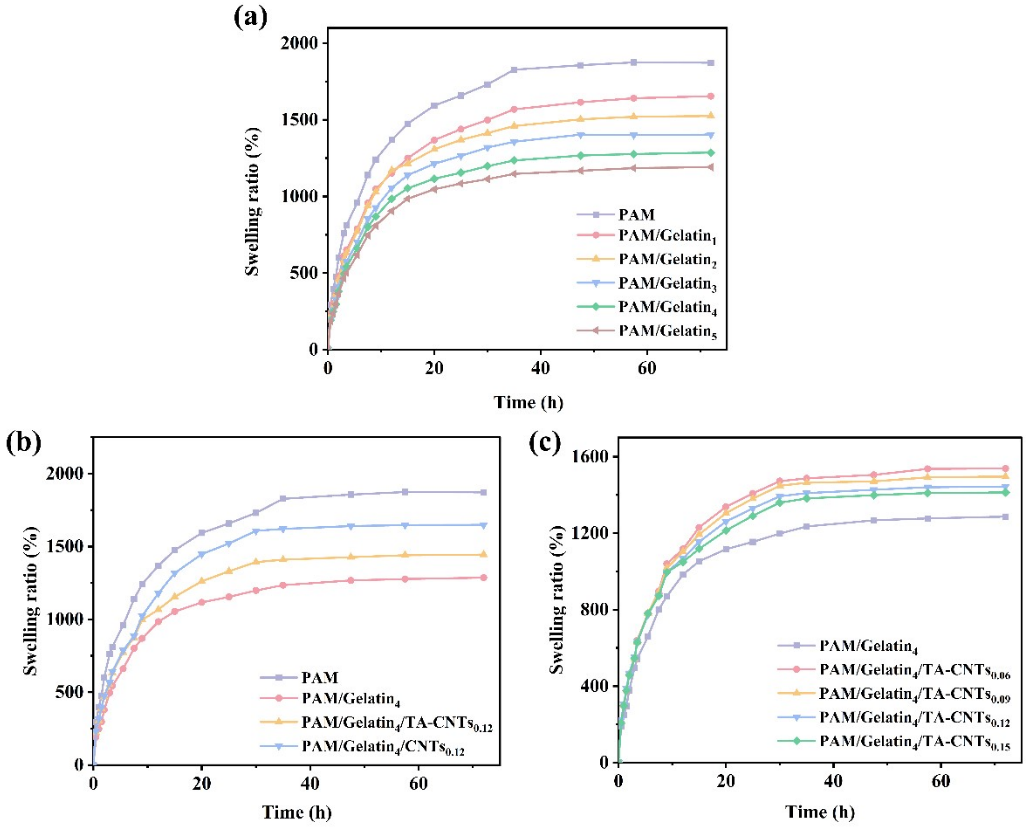

Fig. S5 (a) Swelling rate of PAM, PAM/$Gelatin_1$, PAM/$Gelatin_2$, PAM/$Gelatin_3$, PAM/$Gelatin_4$ and PAM/$Gelatin_5$ hydrogels; (b) Swelling rate of PAM, PAM/$Gelatin_4$, PAM/$Gelatin_4$/TA-$CNTs_{0.12}$ and PAM/$Gelatin_4$/$CNTs_{0.12}$ hydrogels; (c) Swelling rate of PAM/$Gelatin_4$ and PAM/$Gelatin_4$/TA-CNTs hydrogels with TA-CNTs content as variable.

The swelling ratio of PAM, PAM/Gelatin, PAM/$Gelatin_4$/CNTs and PAM/$Gelatin_4$/TA-CNTs hydrogels was shown in Fig. S5. It can be seen from Fig. S5 (a) that the swelling rate of PAM single network hydrogel is the largest, and the swelling rate of PAM/Gelatin double network hydrogel decreases with the increase of gelatin content. This is because the double network structure is formed after adding gelatin. The more gelatin is added, the denser the network structure is, and the swelling rate decreases. From Fig.S5 (b), it can be seen that the swelling rate of the hydrogel after adding CNTs is larger than that of PAM/$Gelatin_4$ double network hydrogel. On the one hand, there are a small amount of oxygen-containing groups on the surface of CNTs. On the other hand, CNTs are dispersed unevenly in the hydrogel matrix, destroying the original dense double network structure, thereby increasing the water absorption of the hydrogel. After adding TA-CNTs, the rich oxygen-containing groups of TA increase the hydrophilicity of the hydrogel and allow more water molecules to enter the hydrogel, but at the same time, TA-CNTs can generate hydrogen bond interaction with the hydrogel matrix and act as a nano-crosslinking point, which increases the crosslinking density of the hydrogel and inhibits the expansion of the hydrogel. With the increase of TA-CNTs content, the number of cross-linking points of the double network hydrogel increases, resulting in a denser network structure, thereby reducing the swelling rate (Fig.S5 (c)).

Table S1. Comparisons between the PAM/Gelatin/TA-CNTs hydrogel sensor and reported hydrogel sensor.

Composition	GF	Strain (%)	Reference
PAM/Gelatin/PDA-CNTs	3.01	200	[1]
PAM/SA/TA-CNTs	4.38	400	[2]
PEDOT: PSS/GO/ PNIPAM	4.96	800	[3]
HPAAN/PDA	0.84	200	[4]
PVA/CMC/TA/MXene	2.9	700	[5]
PANI/PVA/CBA	1.71	300	[6]
PDA-rGO/CMCNa/PAM	6.44	500	[7]
PAM/Gelatin/TA-CNTs	5.50	500	This work

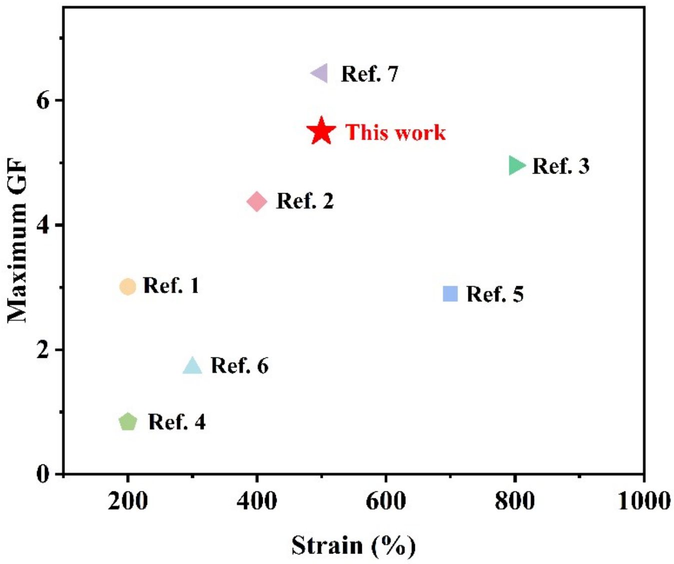

Fig. S6 GF value comparison with different hydrogel sensors.

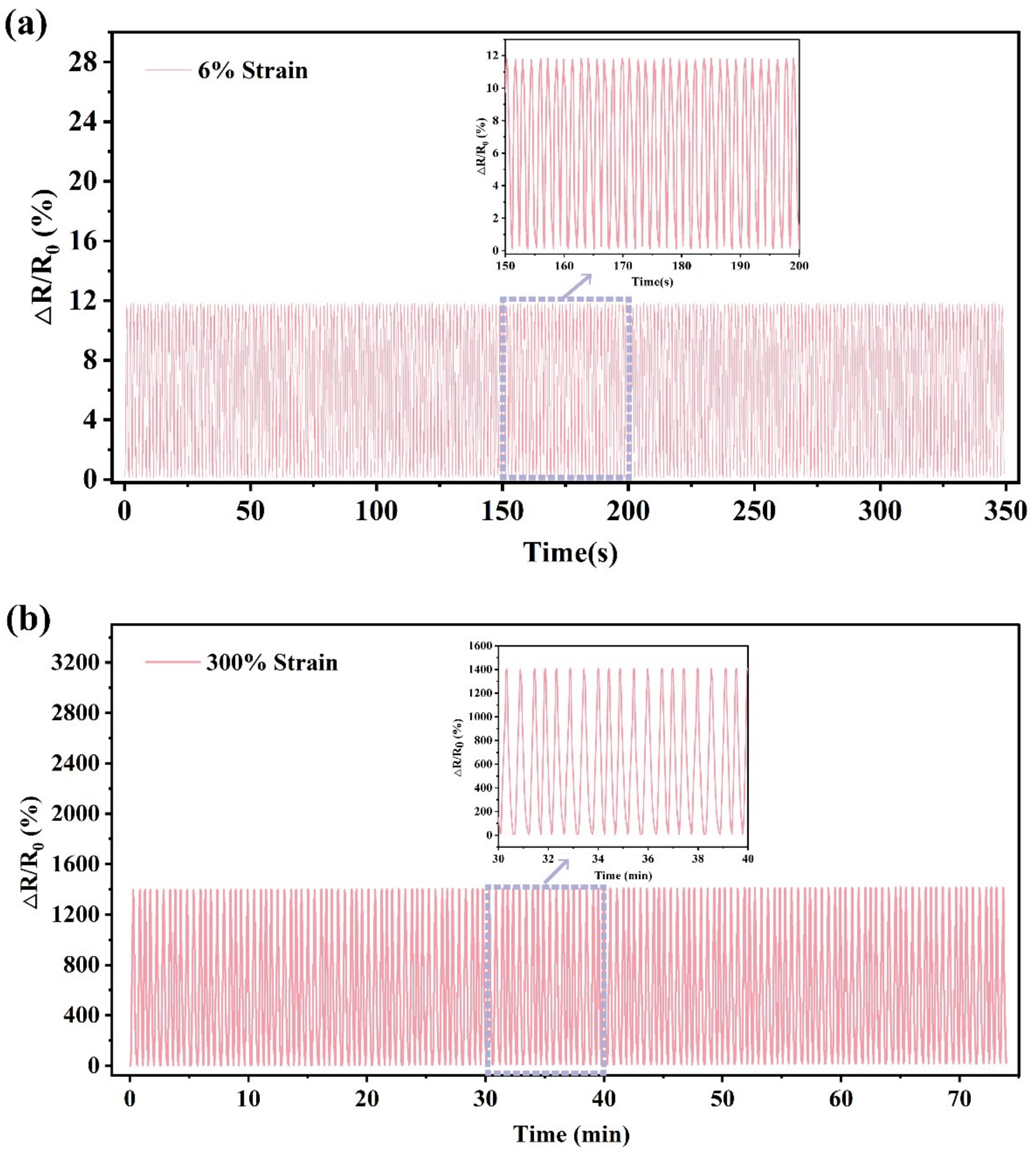

Fig. S7 (a) Relative resistance change after 250 cycles of 6 % strain cycling; (b) Relative resistance change after 150 cycles of 300 % strain cycling.

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