Support Information

Phytic acid/Tannic acid reinforced hydrogels with ultra-high strength for human motion monitoring and arrays

Jiegao Xie[a], Yafei Qin*[a], Yu Zeng[a], Ruibo Yuan[a], Xinyu Lu[a], Xiaojing Yang[a], Erjiong Wei[a], Chenkai Cui[a]

Table S1. Composition of P/G/A_X/T hydrogel

PVA(g)	Gly(g)	PA(g)	TA	DI(g)
1	1	0.5	10wt% solution	7.5
		1		7
		1.5		6.5
		2		6

Table S2. Composition of P/G_X/A/T hydrogel

PVA(g)	Gly(g)	PA(g)	TA	DI(g)
1	0.5	1	5wt% solution	7.5
	1			7
	1.5			6.5
	2			6

Table S3. Composition of P/G/A/T_X hydrogel

PVA(g)	Gly(g)	PA(g)	TA	DI(g)
1	1.5	1	5wt% solution	6.5
			10wt% solution	
			15wt% solution	
			20wt% solution	

[a] *Faculty of Mechanical and Electrical Engineering, Kunming University of Science and technology, Jing-ming,727, Yun nan Province, People's Republic of China. qinyafei_kmust@foxmail.com*

Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/x0xx00000x

Table S4. Comparison of wearable sensor based on P/G/A/T hydrogels in this work with reported hydrogel sensors

Hydrogel	Elongation(%)	Strength(MPa)	GF[a]	Response time(ms)	Repeatability	Antibacterial	Ref.
PVA-CNF	660	2.1	1.5	NA	500 cycles	NA[b]	[1]
PGN	570.7	3.1	4.01	400	300 cycles	NA	[2]
P(AA-MEA)-CS-Fe	1199	0.462	5.25	310	100 cycles	NA	[3]
PGA	472	0.28	2.14	230	540 cycles	NA	[4]
PAA-[EMIm][DCA]	>850	0.0363	1.29	200	>1400 cycles	NA	[5]
PAM/SA/Ca^{2+}	>1700	0.385	NA	800	200 cycles	NA	[6]
PAM/CS-PA	>1000	0.1505	8.24	291	200 cycles	Yes	[7]
PVA/Gly/PA/TA	>1000	9.341	4.76	364	500 cycles	Yes	This work

[a] The GF between 0%~300% stain was selected as comparison.

[b] NA:not applicable.

Fig. S1. The brightness change of LED lights follows the stretching

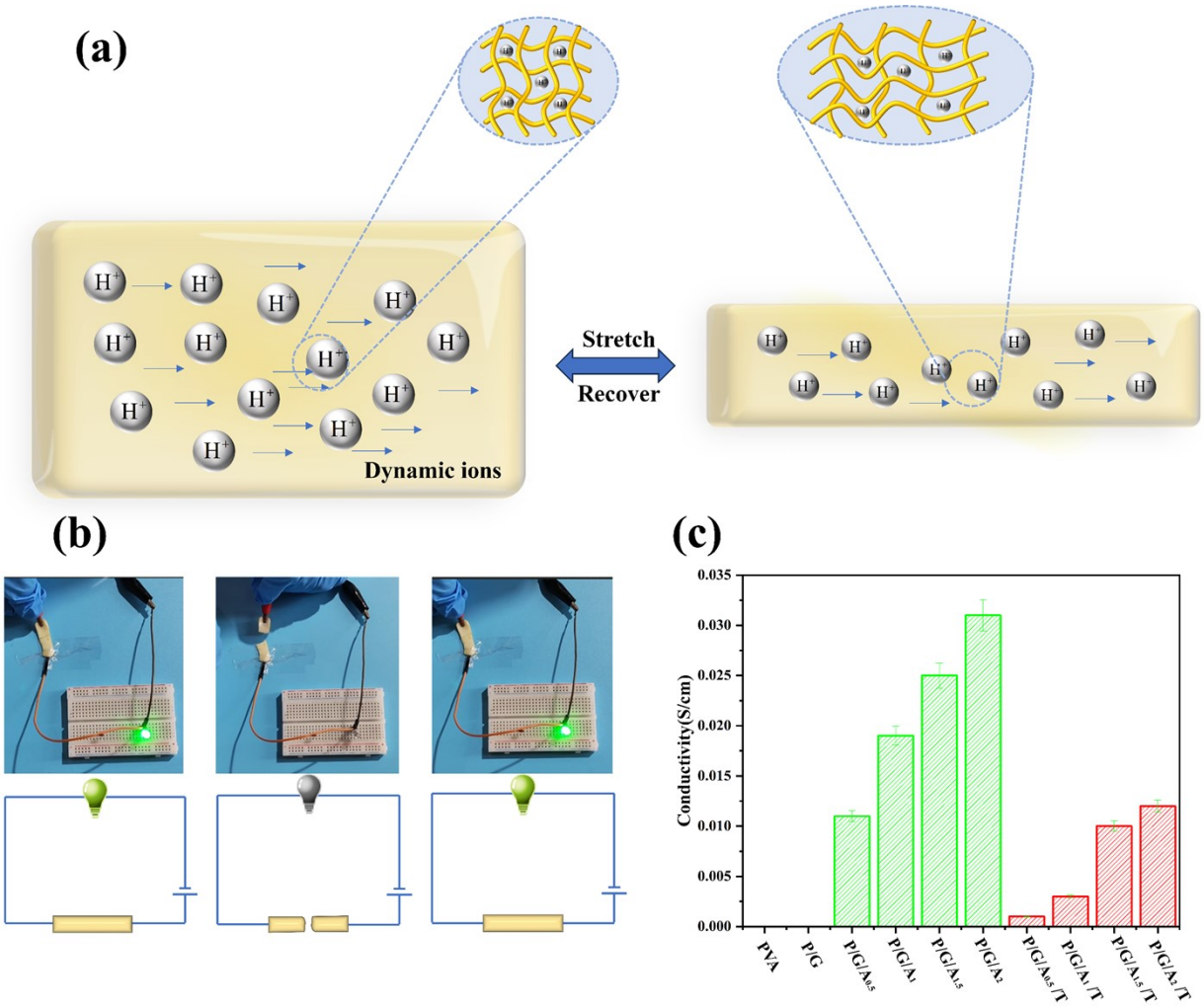


Fig. S2. conducting mechanism diagram of hydrogel

Phytic acid and tannic acid are used as electrolytes. Phytic acid and tannic acid ionize free H^+ in water, and the directed movement of a large number of free H^+ imparts excellent conductivity to the hydrogel. As shown in Fig S2 (a), with the stretching of the hydrogel, external stress causes the ion channels inside the hydrogel to elongate and narrow, resulting in a decrease in ion transport rate, thus responding to stress-induced changes in resistance signals. As a conductor, the hydrogel can effectively light up an LED. When cut off, the bulb goes out, and when reconnected, the bulb lights up again (Fig S2 (b)). In addition, as shown in Fig S2 (c), PVA and P/G are almost non-conductive. Meanwhile, we found that the overall conductivity of the hydrogel without soaking in tannic acid solution is higher than that of the hydrogel soaked in tannic acid. Specifically, the conductivity of P/G/A_2 hydrogel is 0.031 S/cm, and after soaking in tannic acid solution, its conductivity decreases to 0.012 S/cm. We speculate that this is due to further complexation of the hydrogel, leading to smaller and fewer ion channels, which is unfavorable for ion movement and thus reduces the conductivity of P/G/A_2/T hydrogel.

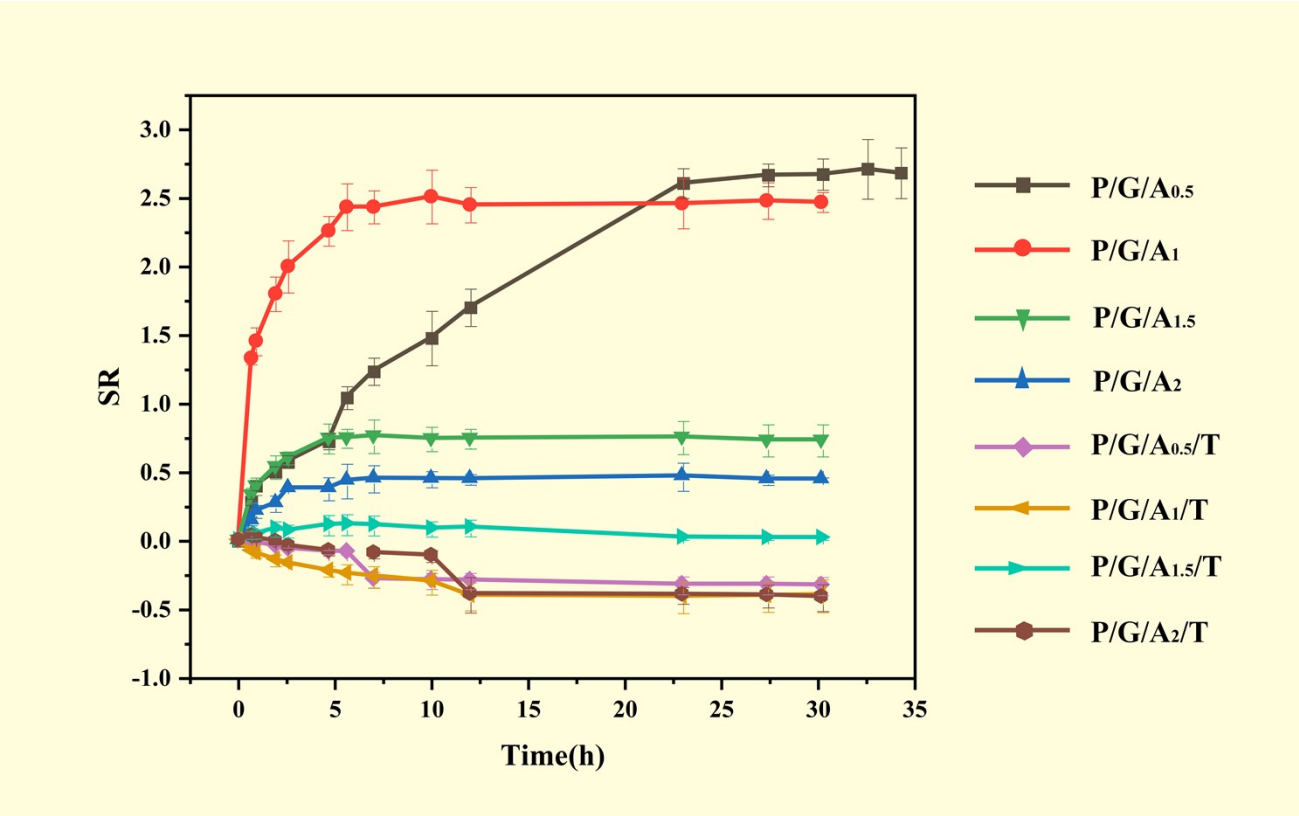


Fig. S3. Swelling ratio of different hydrogels in this work

This study observed degradation in both PVA and P/G in deionized water, with no swelling phenomenon. However, with the introduction of phytic acid, a large number of hydrogen bonds are formed between polymer chains inside the hydrogel, enhancing its resistance to swelling. As shown in Fig S3, the swelling ratio (SR) of P/G/A_2 hydrogel is only 0.448, while the SR of P/G/$A_{0.5}$ hydrogel is close to 2.8, with SR decreasing as the phytic acid content increases. Additionally, the hydrogel soaked in tannic acid solution initially maintains swelling equilibrium or did not have too much swelling behavior, as P/G/A/T hydrogel used for swelling experiment itself was a product of swelling equilibrium.

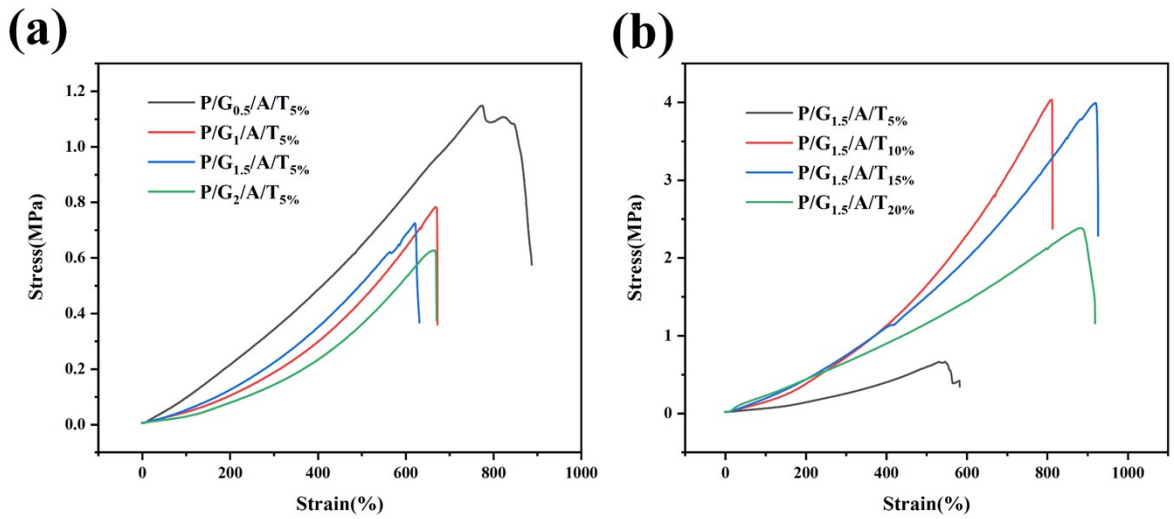
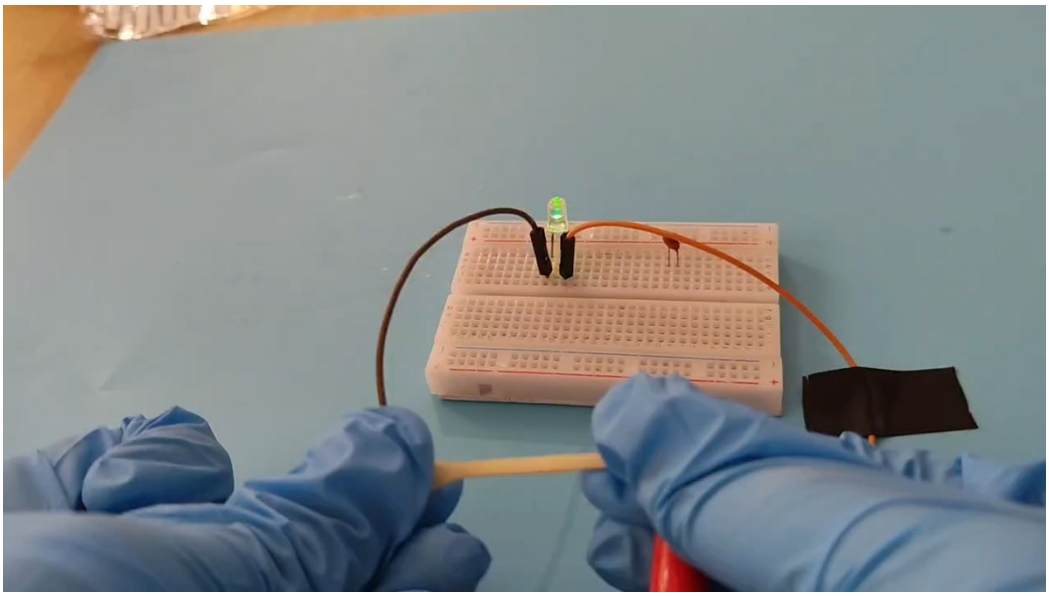
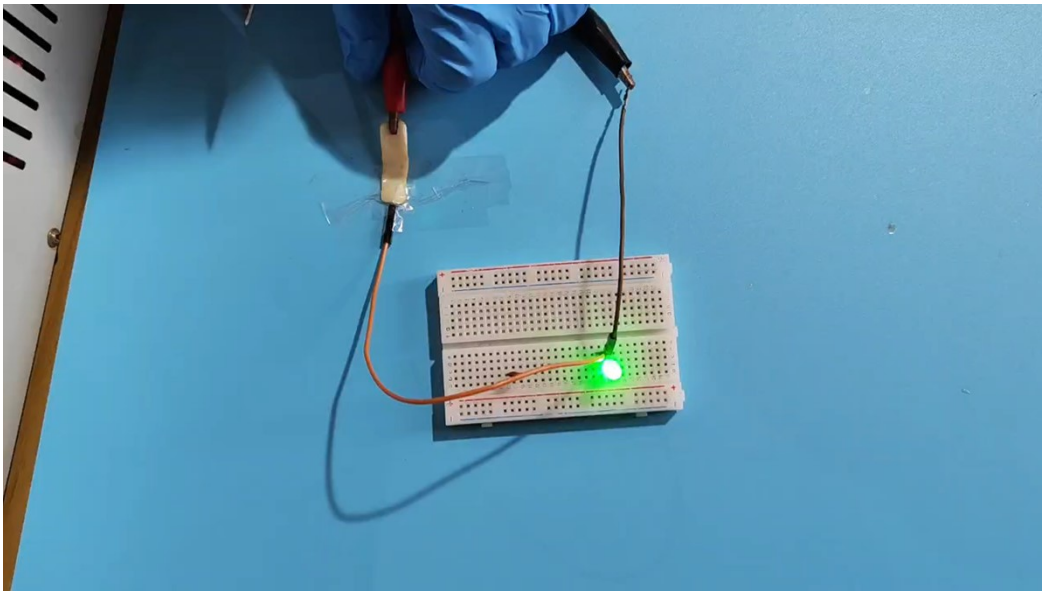


Fig. S4. The effect of different concentrations of glycerol and TA on the mechanical properties of hydrogels

The effects of different concentrations of glycerol and tannic acid on the mechanical properties of P/G/A/T hydrogels were studied. As shown in Fig S4 (a), the tensile strain and stress of $P/G_{0.5}/A/T_{5\%}$ hydrogel reached 829% and 1.10MPa, respectively, which were significantly higher than those of other hydrogels with glycerol concentration. This is because the increase of glycerol concentration interfered with the hydrogen bond cross-linking between PVA chain and additives, thereby reducing the mechanical properties of P/G/A/T hydrogel[8]. Mealwhile, with the increase of tannic acid concentration, the hydrogen bond complexation in P/G/A/T hydrogel becomes more intensive, and the mechanical properties of hydrogel become stronger. Compared with $P/G_{1.5}/A/T_{5\%}$ hydrogel and $P/G_{1.5}/A/T_{15\%}$ hydrogel, their strength and strain increase from 0.665MPa and 548% to 3.99MPa and 922% respectively (Figure S4 (b)). Interestingly, the same situation as in Fig 4 (e) reappears. The mechanical properties of $P/G_{1.5}/A/T_{20\%}$ hydrogel decreased sharply. Obviously, the 20% concentration of TA solution led to the excessive crosslinking of the hydrogel, which led to the decline of the mechanical properties of P/G/A/T hydrogel. Therefore, the optimal TA concentration of the hydrogel during immersion should be 10% or 15%.



Video. S1. The brightness change of LED lights follows the stretching- releasing cycle



Video. S2. Conductivity of hydrogel during cut off-contact cycle

References

1. Y. Ye, Y. Zhang, Y. Chen, X. Han and F. Jiang, *Advanced Functional Materials*, 2020, **30**.
2. S. Pan, M. Xia, H. Li, X. Jiang, P. He, Z. Sun and Y. Zhang, *Journal of Materials Chemistry C*, 2020, **8**, 2827-2837.
3. Z. Zhao, X. Qin, L. Cao, J. Li and Y. Wei, *Int J Biol Macromol*, 2022, **212**, 123-133.
4. C. Hu, Y. Zhang, X. Wang, L. Xing, L. Shi and R. Ran, *ACS Appl Mater Interfaces*, 2018, **10**, 44000-44010.
5. J. Lai, H. Zhou, Z. Jin, S. Li, H. Liu, X. Jin, C. Luo, A. Ma and W. Chen, *ACS Appl Mater Interfaces*, 2019, **11**, 26412-26420.
6. H. Sun, K. Zhou, Y. Yu, X. Yue, K. Dai, G. Zheng, C. Liu and C. Shen, *Macromolecular Materials and Engineering*, 2019, **304**.
7. Q. Zhang, X. Liu, J. Zhang, L. Duan and G. Gao, *Journal of Materials Chemistry A*, 2021, **9**, 22615-22625.
8. Y. Peng, B. Yan, Y. Li, J. Lan, L. Shi and R. Ran, *Journal of Materials Science*, 2020, **55**, 1280-1291.