## Tough and adhesive conductive hydrogel with fast gelation from polyphenols-aluminium ion dual self-catalysis system for wearable strain sensors and triboelectric nanogenerators

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Hydrogel	Tensile strengt h (kPa)	Tensil e strain (%)	Adhesion (kPa)	GF <sup>a)</sup>	Ref
AgNP@GCOL/PAA	123	916	42.8	N.D.	[S1]
MoS <sub>2</sub> -Containing Composite	30	85	No	N.D.	[S2]
PAA/ZnCl <sub>2</sub> /lignin	2510	336	62.05	1.678 (0-10%) 2.104 (10-300%)	[S3]
P(AA-DAM)/PVA	50	1050	17.42	1.24 (0-50%) 3.57 (50-300%)	[S4]
PAM@CNC/TA-Ag NC	128.12	1500	17	1.02 (0-200%)	[85]
AL-Cu@W/EG-PAM	136.1	550	26.8	N.D.	[S6]
PVA/PHEAA-TA- Al <sup>3+</sup>	240	920	108	1.3 (0-200%) 2.7 (200-600%)	This work

 Table S1. Summary of mechanical properties of the hydrogels by self-catalytic system.

<sup>a)</sup>N.D.: no date.



**Fig. S1** (a) Discoloration mechanism diagram of Coomassie brilliant blue as an indicator of hydroxyl radical, (b) The color change of BB was measured by spectrophotometry under different conditions. (c) ESR spectrum of precursor solution of PVA/PHEAA-TA-Al<sup>3+</sup> gel.



**Fig. S2** High-resolution XPS spectra of (a) C 1s and (b) Al 2p of PVA/PHEAA-TA-Al<sup>3+</sup> gel.



**Fig. S3** The loading-unloading cyclic curve of PVA/PHEAA-TA-Al<sup>3+</sup> DN gel under (a) 200% strain and (b) the relative recovery rate of each cycle. (c) The consecutive compressive cyclic loading-unloading curves of PVA/PHEAA-TA-Al<sup>3+</sup> DN gel under 80% strain and (d) relative recovery rate of each cycle.



**Fig. S4** Visualization of the self-healing ability of PVA/PHEAA-TA-Al<sup>3+</sup> DN gel for 6 h, as well as (a) the stretching and (b) bending photos of the self-healing hydrogel. (c) Illuminance variation of LED light during PVA/PHEAA-TA-Al<sup>3+</sup> DN gel cutting/healing, (d) The stress-strain curve of PVA/PHEAA-TA-Al<sup>3+</sup> DN gel after healing for 6 h at 25°C.



Fig. S5 The adhesive (a) curves and (b) strength of the hydrogels on different substrates.



**Fig. S6** Durability test of PVA/PHEAA-TA-Al<sup>3+</sup> gel-based sensor for 500 cycles from 0% to 50% strain.



Fig. S7 The values of  $V_{OC}$  under different (a) frequencies (0.5-2.5 Hz) and (b) pressures (5-40 N).

## References

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