International Journal of Bioprinting

RESEARCH ARTICLE

3D printing of mechanically tough and self-healing hydrogels with carbon nanotube fillers

Supplementary File

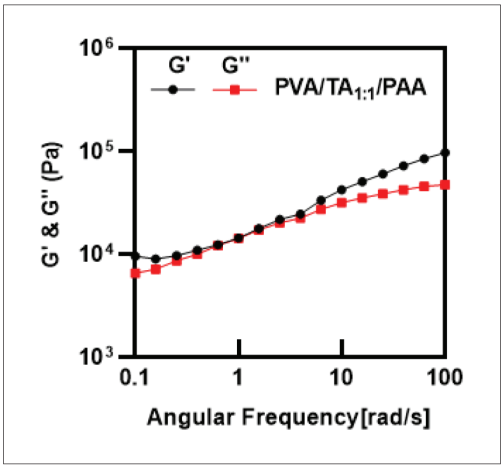

Figure S1. Rheological data of G′ and G″ as a function of angular frequency for PVA/$TA_{1:1}$/PAA hydrogel ink.

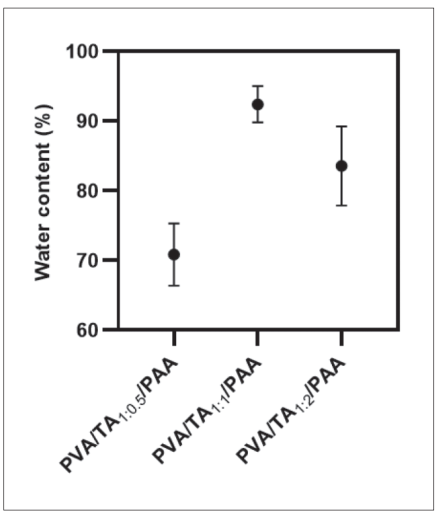

Figure S2. The water content of various mass ratios of PVA/TA/PAA hydrogel ink.

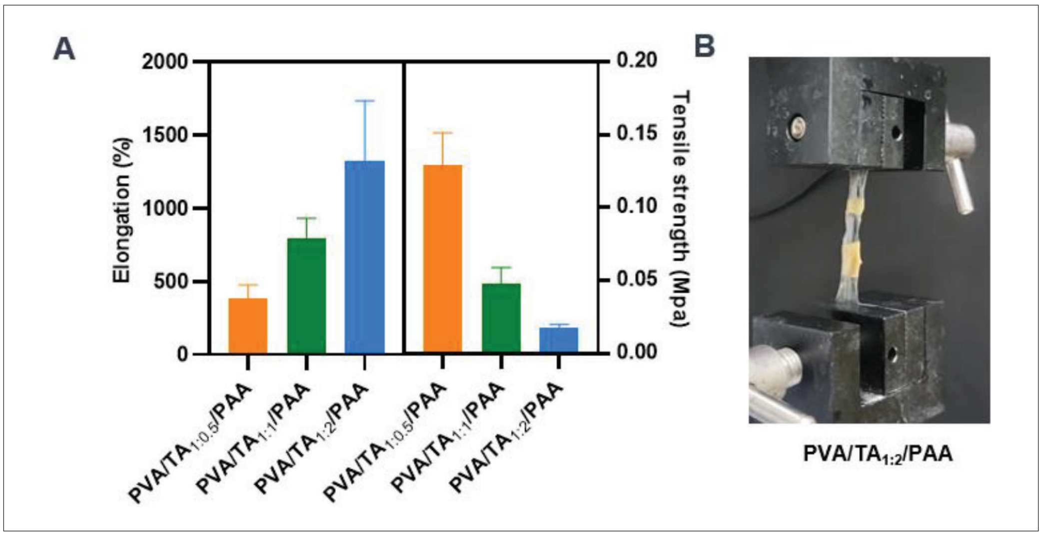

Figure S3. (A) Mechanical properties of various ratios of printed hydrogel inks. (B) Optical photograph mechanical property of printed PVA/$TA_{1:2}$/PAA hydrogel.

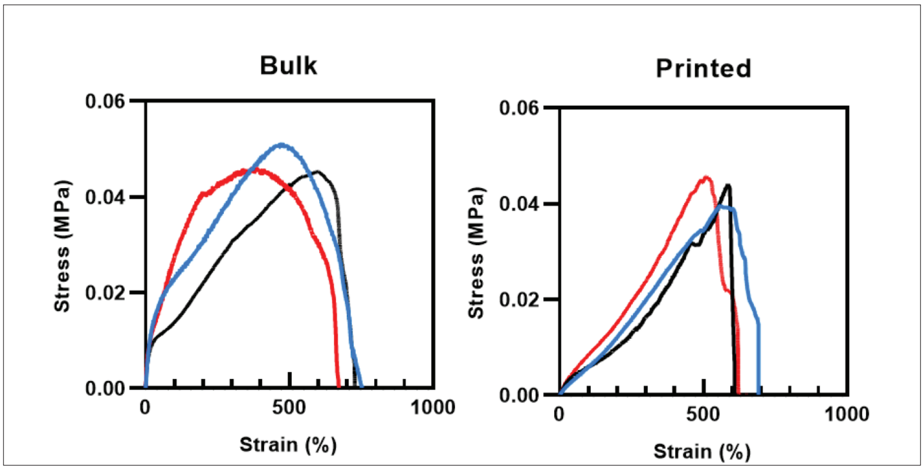

Figure S4. Strain–stress curves of bulk and printed PVA/$TA_{1:1}$/PAA hydrogel ink.

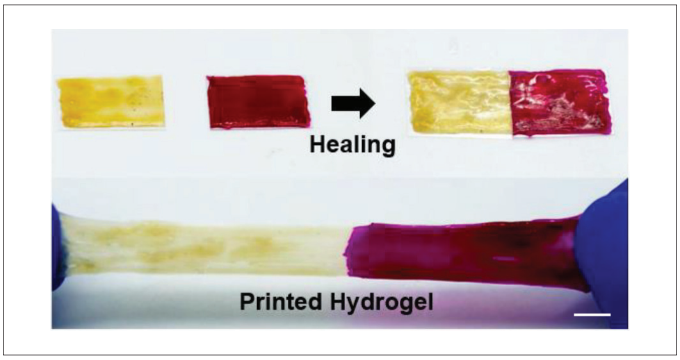

Figure S5. Photographs of printed PVA/$TA_{1:1}$/PAA hydrogel self-healing hydrogel. Scale bar: 7 mm.

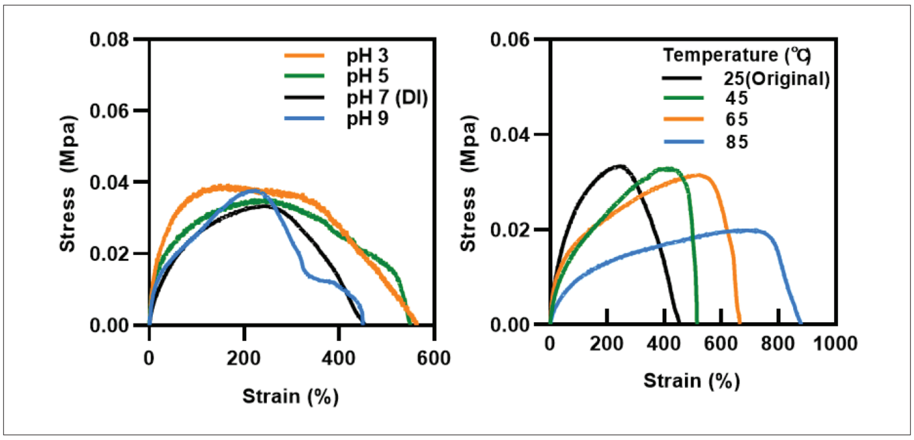

Figure S6. Strain–stress curves of bulk PVA/$TA_{1:1}$/PAA hydrogel ink with varying pH conditions and temperature.

Table S1. A comparison table of recently reported multi-functional hydrogels for bioelectronics.

Material	Strength/kPa	Elongation/%	Self-healing ability	Printability	Conductivity	Ref
2-ureido-4[1H]-pyrimidinone (UPy) and polyaniline/poly(4-styrenesulfonate) (PANI/PSS)	Not reported	670	Yes	Resolution of ~1.2 mm but not 3D-printed	13 S/m and GF of 3.4	[1]
Cassava starch, boric-acid, and rubber latex	1010	1500	Yes (≈72% efficiency in 90 min)	3D printable (resolution was not reported)	GF of 2.027	[2]
Halloysite nanotube (HNT), polydopamine (PDA), PVA, and ferric ions (Fe^{3+})	140–560	30,000	Yes (≈99% efficiency in a 360 s)	Resolution of ~1.5 mm nozzle and 3D printable	0.005–0.01 S/m and GF of 2.6	[3]
PANI and PAA with phytic acid	500–2000	500	Yes (≈99% efficiency in a 24 h)	Not reported	12 S/m (GF was not reported)	[4]
PVA, TA, PAA, and CNT	45.6	650	Yes (≈86% efficiency in a 300 s)	Resolution of ~100 μm and 3D printable	0.3–1 S/m and GF of 4.457	This work

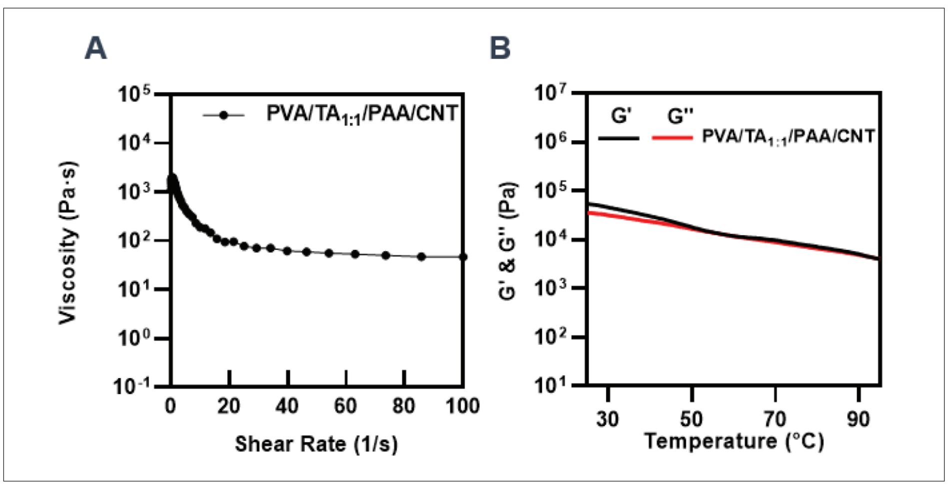

Figure S7. Rheological property of (A) viscosity as a function of shear rate. (B) Storage modulus (G') and loss modulus (G") as a function of temperature.

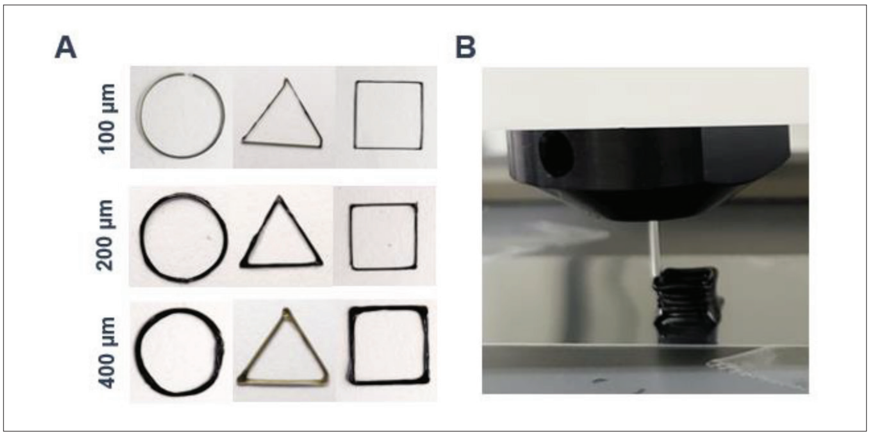

Figure S8. Printability of PVA/$TA_{1:1}$/PAA/CNT hydrogel ink. (A) 2D-printing performance of various shapes through 400-, 200-, and 100-µm diameter nozzles. (B) 3D-printed hydrogel by stacked structure.

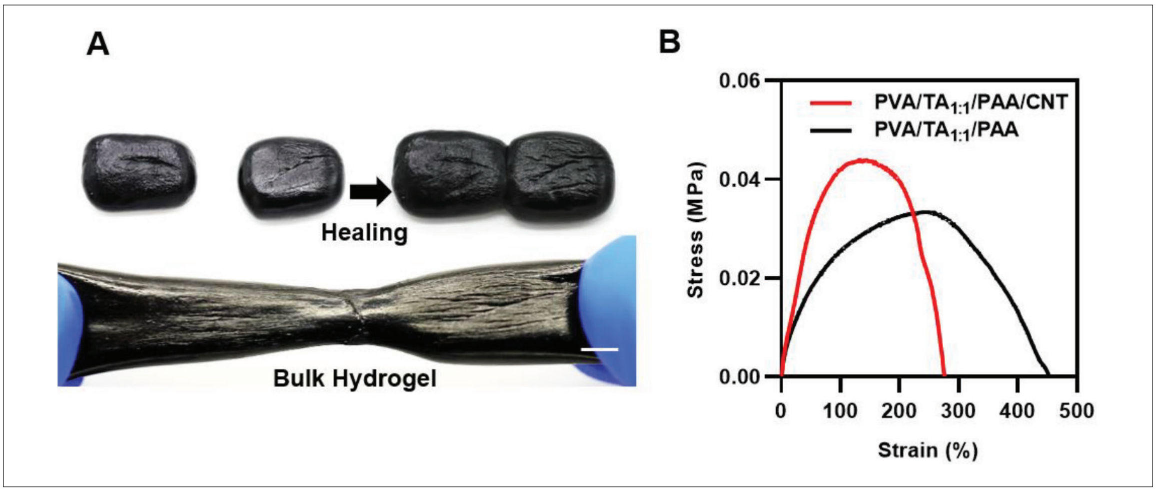

Figure S9. (A) Photographs of bulk PVA/$TA_{1:1}$/PAA/CNT hydrogel self-healing hydrogel (scale bar: 7 mm). (B) Strain–stress curves of PVA/$TA_{1:1}$/PAA hydrogel and PVA/$TA_{1:1}$/PAA/CNT hydrogel ink after self-healing for 180 s.

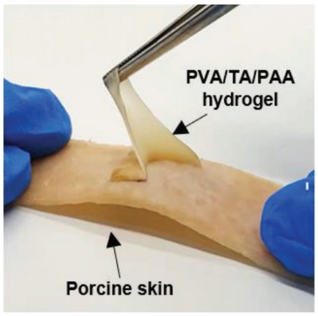

Figure S10. Photographs of PVA/TA/PAA hydrogel adhesion on porcine skin.

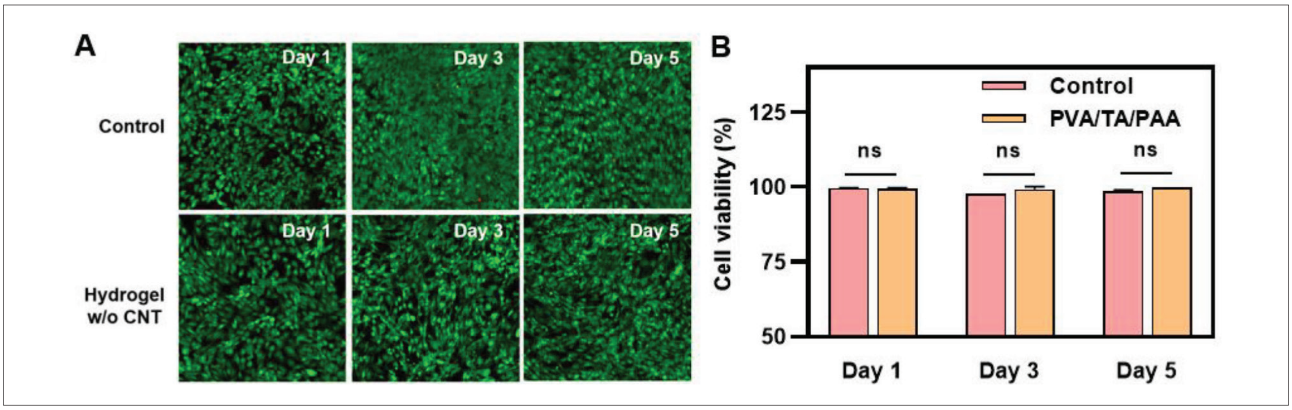

Figure S11. (A) Fluorescent images of *in vitro* biocompatibility test after 1, 3, and 5 days. (B) The percentage of cell viability of *in vitro* biocompatibility test of the hydrogel (ns: no significant differences; $n = 3$; n is the sample size for each group).

Supplementary references

S1. Chen J, Peng Q, Thundat T, *et al.*, 2019, Stretchable, injectable, and self-healing conductive hydrogel enabled by multiple hydrogen bonding toward wearable electronics. *Chem Mater*, 31: 4553–4563.

http://doi.org/10.1021/acs.chemmater.9b01239

S2. Zhao W, Huang B, Zhu L, *et al.*, 2022, Printable hydrogels based on starch and natural rubber latex with high toughness and self-healing capability. *Int J Biol Macromol*, 218: 580–587.

http://doi.org/https://doi.org/10.1016/j.ijbiomac.2022.07.148

S3. Karolina Pierchala M, Kadumudi FB, Mehrali M, *et al.*, 2021, Soft electronic materials with combinatorial properties generated via mussel-inspired chemistry and halloysite nanotube reinforcement. *ACS Nano*, 15: 9531–9549.

http://doi.org/10.1021/acsnano.0c09204

S4. Wang T, Zhang Y, Liu Q, *et al.*, 2018, A self-healable, highly stretchable, and solution processable conductive polymer composite for ultrasensitive strain and pressure sensing. *Adv Funct Mater*, 28: 1705551.

http://doi.org/https://doi.org/10.1002/adfm.201705551