Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting Information

Multi-Interaction Conductive Double-Network Polyelectrolyte Hydrogel with High Stretchability, Self-Adhesion, and Tunable Transparency for Bioelectronic Sensing and Information Encryption

Dongdong Lu[a,b*#], Zilong Zhu[a#], Mingning Zhu[c], Peng Zhang[d], Xiaodong Xiang[a,d *]

a. Department of Materials Science and Engineering, Southern University of Science and Technology, Shenzhen, Guangdong, 518000, P. R. China

b. Dongguan Key Laboratory of Interdisciplinary Science for Advanced Materials and Large-Scale Scientific Facilities, School of Physical Sciences, Great Bay University, Dongguan, Guangdong, 523000, P. R. China

c. School of Biomedical Engineering, Guangdong Medical University, Dongguan 523808, P. R. China

d. Academy for Advanced Interdisciplinary Studies, Southern University of Science and Technology, Shenzhen 518055, P. R. China

*Corresponding authors:

E-mail: ludongdong@gbu.edu.cn

E-mail: xiangxd@sustech.edu.cn

[#]Contributed equally to this work

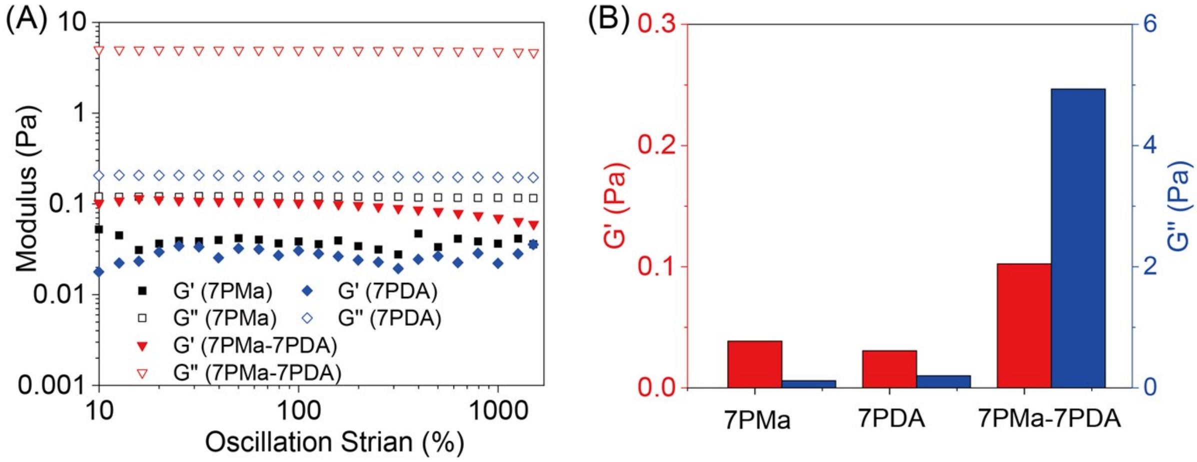

Figure S1. (A) The rheological strain of the 7PMa, 7PDA, and 7PMa-7PDA dispersions. **(B)** G' and G'' obtained from (A) at strain of 100%.

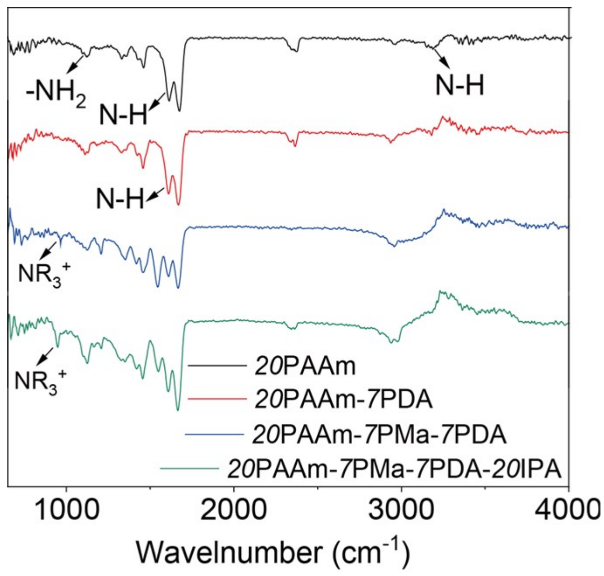

Figure S2. FTIR spectra of *x*PAAm-*y*PMa-*y*PDA-*z*IPA hydrogels.

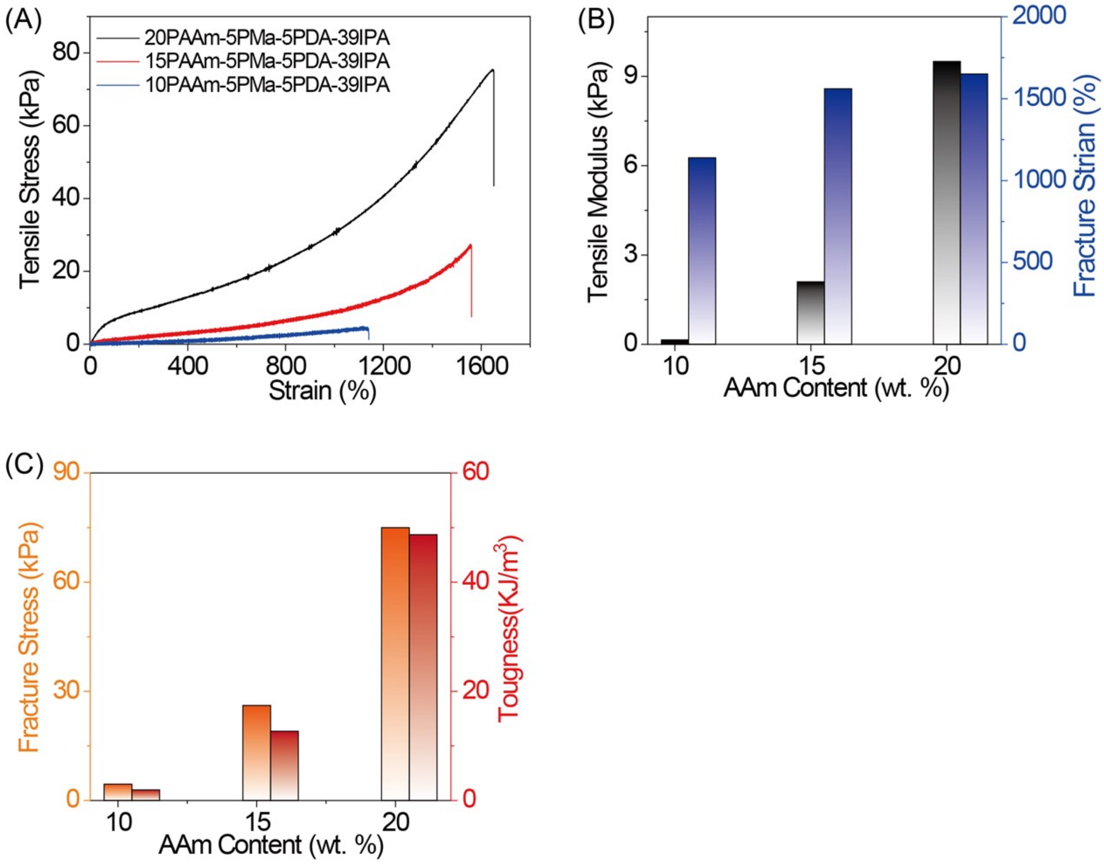

Figure S3. (A) Tensile stress-strain curves of the *x*PAAm-*5*PMa-*5*PDA-*39*IPA hydrogels with different PAAm content. **(B)** The calculated tensile modulus and fracture strain of hydrogels from (A). **(C)** The calculated fracture stress and toughness hydrogels from (A).

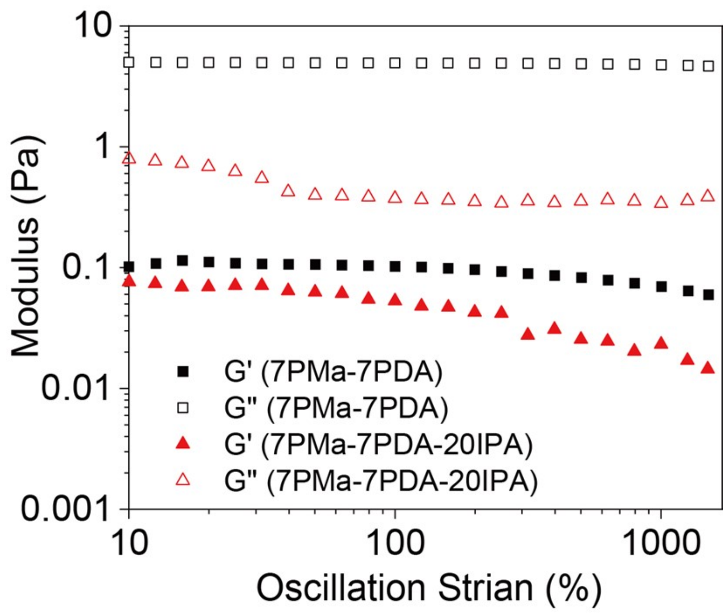

Figure S4. (A) Rheological strain of the 7PMa-7PDA and 7PMa-7PDA-20IPA dispersions.

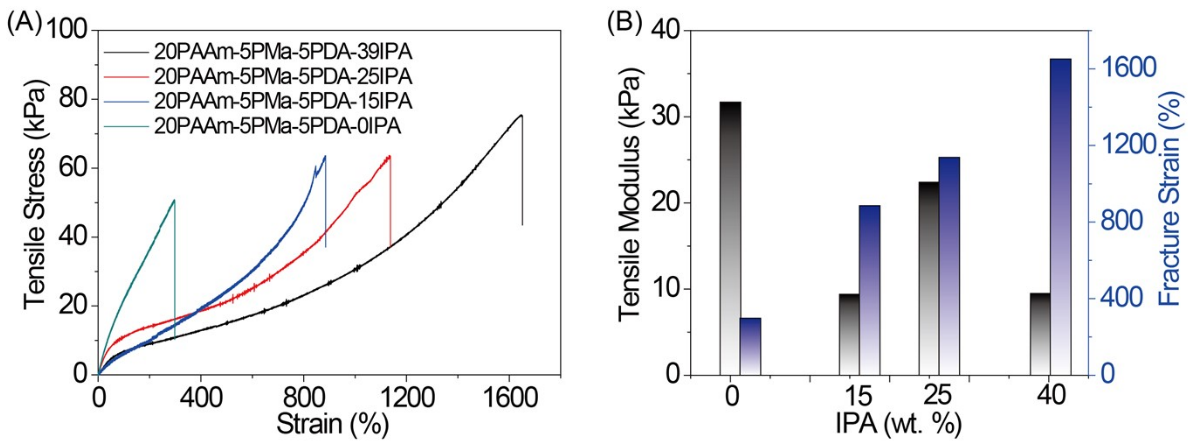

Figure S5. (A) Tensile stress-strain curves of the *20*PAAm-*5*PMa-*5*PDA-*z*IPA hydrogels with different IPA content. **(B)** The calculated tensile modulus and fracture strain of hydrogels from (A).

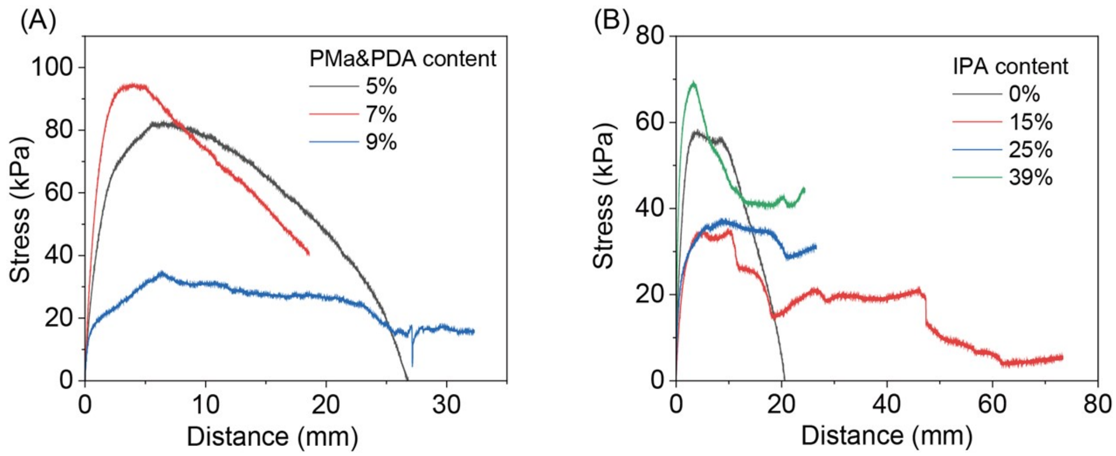

Figure S6. The adhesive curve of the *20*PAAm-*y*PMa-*y*PDA-*0*IPA hydrogels with various polyelectrolytes content **(A)** and the 2*0*PAAm-*5*PMa-*5*PDA-*z*IPA hydrogels with different IPA content **(B)**.

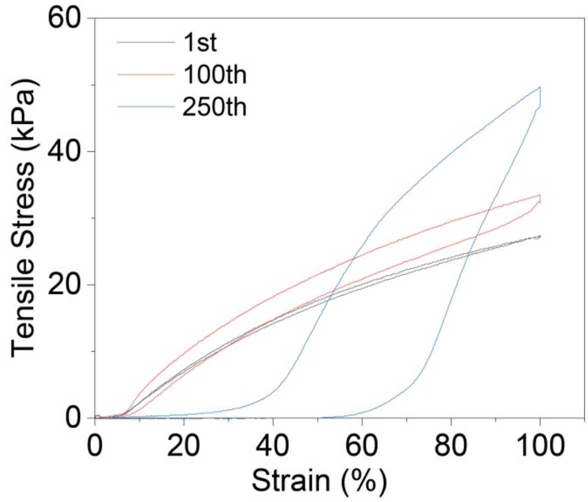

Figure S7. Successive tensile tests with a strain of 100%.

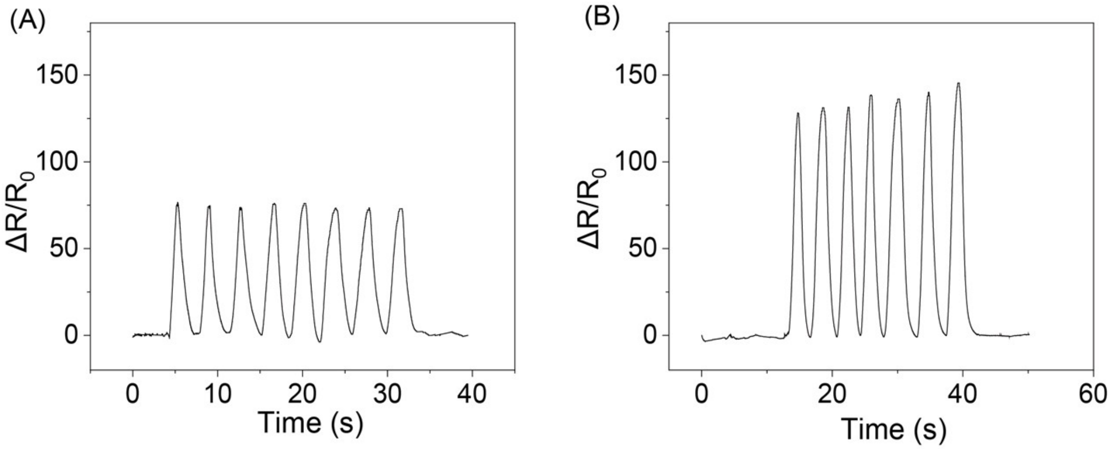

Figure S8. Electronic signal investigation under 100% strain underwater **(A)** and at 40 °C **(B)**.

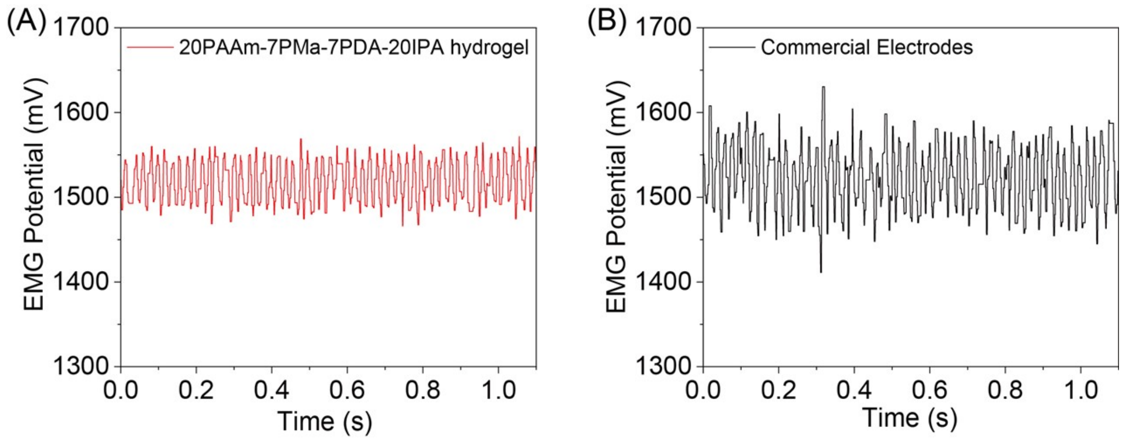

Figure S9. The signal-to-noise ratio (SNR) of 2*0*PAAm-*7*PMa-*7*PDA-*10*IPA hydrogels **(A)** and commercial electrodes **(B)**.

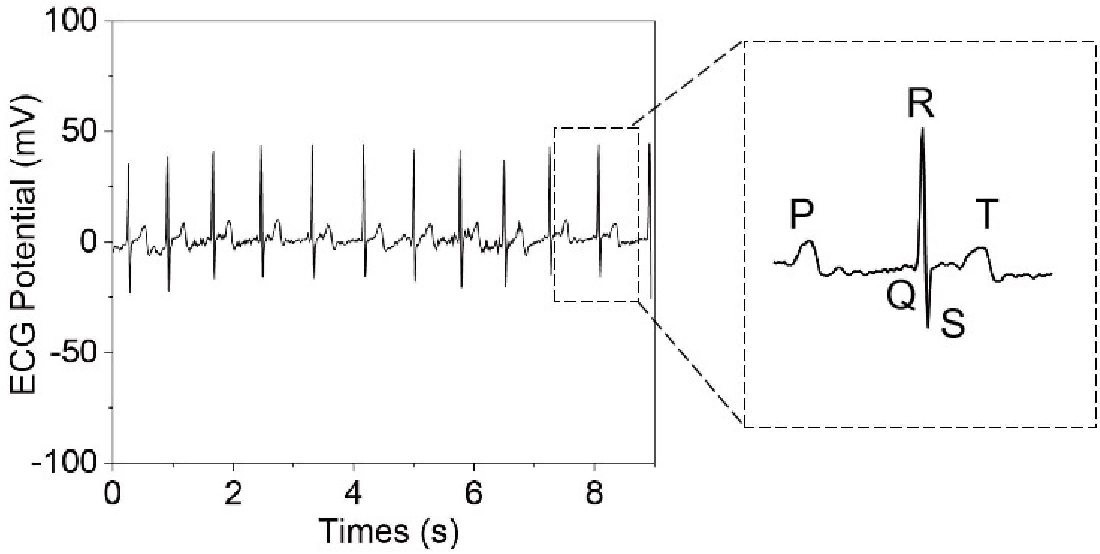

Figure S10. The ECG signals collected from the 2*0*PAAm-*7*PMa-*7*PDA-*10*IPA hydrogels electrolyte after being reposted 10 times during rest.

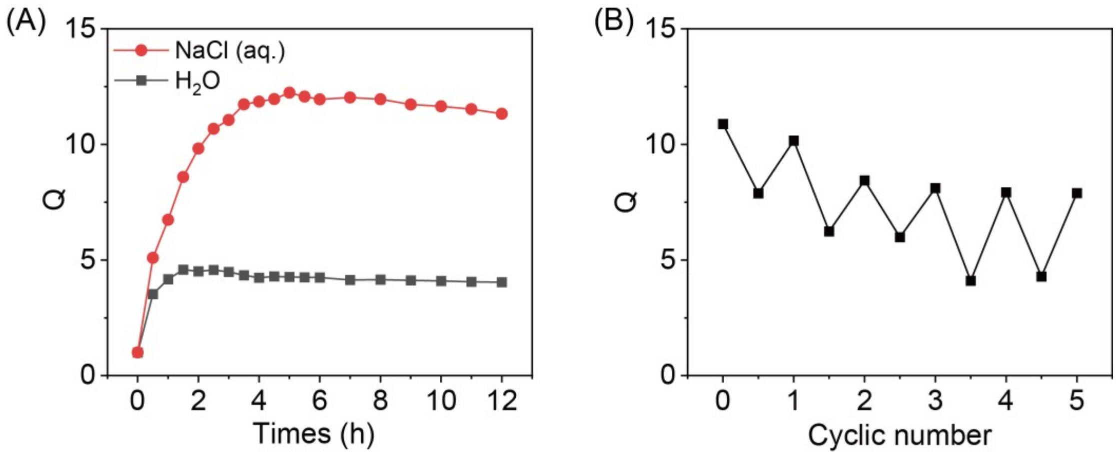

Figure S11. (A) The swelling kinetics of 2*0*PAAm-*7*PMa-*7*PDA-*20*IPA hydrogels immersed in water and aqueous NaCl solution. **(B)** Swelling ratio variation of 2*0*PAAm-*7*PMa-*7*PDA-*20*IPA hydrogels cyclically swollen in water and aqueous NaCl.

Table S1. Summary of the DN polyelectrolytes hydrogel's components used for HTT and the tensile testing results.

DN polyelectrolytes hydrogel	AAm (wt.%)	PMMA Na (wt.%)	PDDA (Mn<10W) (wt.%)	Solvent (wt.%)	Tensile modulus (kPa)	Fracture strain (%)
*10*PAAm-*5*PMa-*5*PDA-*39*IPA	10	5	5	39	0.15	1139
*15*PAAm-*5*PMa-*5*PDA-*39*IPA	15	5	5	39	2.1	1560
*20*PAAm-*5*PMa-*5*PDA-*39*IPA	20	5	5	39	9.5	1651
*20*PAAm-*5*PMa-*5*PDA-*25*PA	20	5	5	25	22.4	1137
*20*PAAm-*5*PMa-*5*PDA-*15*IPA	20	5	5	15	9.4	885
*20*PAAm-*7*PMa-*7*PDA-*39*IPA	20	7	7	39	2.5	1918
*20*PAAm-*7*PMa-*7*PDA-*20*IPA	20	7	7	20	10.8	1036
*20*PAAm-*7*PMa-*7*PDA-*0*IPA	20	7	7	0	37.1	404
*20*PAAm-*0*PMa-*0*PDA-*0*IPA	20	0	0	0	31.3	510
*20*PAAm-*0*PMa-*7*PDA-*0*IPA	20	0	7	0	36.4	440
*20*PAAm-*5*PMa-*5*PDA-*0*IPA	20	5	5	0	36.3	280
*20*PAAm-*9*PMa-*9*PDA-*0*IPA	20	9	9	0	38.1	280
*20*PAAm-*9*PMa-*9*PDA-*15*IPA	15	9	9	15	14.3	500
*20*PAAm-*9*PMa-*9*PDA-*0*IPA	15	9	9	0	33.1	440
DN polyelectrolytes hydrogel	AAm (wt.%)	PMMA Na (wt.%)	PDDA (Mn10-20W) (wt.%)	Solvent (wt.%)	Tensile modulus (kPa)	Fracture strain (%)
*20*PAAm-*5*PMa-*5*PDA-	20	5	5	39	12.0	1078

*39*IPA						
*20*PAAm-*5*PMa-*5*PDA-*0*IPA	20	5	5	0	34.4	300
*20*PAAm-*7*PMa-*7*PDA-*0*IPA	20	7	7	0	28.5	440
*20*PAAm-*9*PMa-*9*PDA-*0*IPA	20	9	9	0	47.1	200
DN polyelectrolytes hydrogel	AAm	PMMA Na	PDDA (Mn>40W)	Solvent (wt.%)	Tensile modulus (kPa)	Fracture strain (%)
*15*PAAm-*9*PMa-*9*PDA-*0*IPA	15	9	9	0	25.0	400
*15*PAAm-*9*PMa-*9*PDA-*15*IPA	15	9	9	15	12.5	460
*20*PAAm-*5*PMa-*5*PDA-*15*IPA	20	5	5	15	20.3	750
*20*PAAm-*5*PMa-*5*PDA-*0*IPA	20	5	5	0	54.2	225
*20*PAAm-*7*PMa-*7*PDA-*0*IPA	20	7	7	0	34.5	240
*20*PAAm-*9*PMa-*9*PDA-*0*IPA	20	9	9	0	64.6	220

Table S2. Comparisons of properties and functional conductive hydrogels.

Composition	Conductive material	Maximum *GF*	Detection window (%)	Adhesive strength (kPa)	Solvent-tunable transparency	Refs.
PAE/LM-Fe	MXene and Fe^{3+}	2.8	0–947	N/A	N/A	1
PAM-Q-M	MXene	2.24	0–1465	N/A	N/A	2
PAM/SA/MXene	MXene	1.40	0–2000	N/A	N/A	3
LMCH hydrogel	MWCNT	10.76	0–200	15.3	N/A	4
PVA/PAM/NaCl	NaCl	24.9	0-950	N/A	N/A	5
LSN-Fe/PAM	Fe^{3+}	1.22	0-90	12	N/A	6

P(NaSS-*co*-DMAEA-Q-*co*-UM)	P(NaSS-*co*-DMAEA)	1.0	0-250	5.5	Yes	7
PVA-PAANa-PAH	PAANa	1.64	0-500	4.7	Yes	8
PAAm-PMa-PDA-IPA	PMa-PDA	3.77	0-1000	37.8	Yes	This work

PAE: PAAc in water-retaining agent (EG); LM: lignin nanosphere-modified; MXene; PAM and PAAm: Polyacrylamide; Q: quaternized cellulose nanofibrils; SA: sodium alginate; MWCNT: multi-walled carbon nanotubes; PVA: polyvinyl alcohol; LSNs: sulfonated lignin-coated silica nanoparticles; PAANa: poly(sodium acrylate); PAH: polyallylamine; DMAEA: N,N-(dimethylamino)ethylacrylate; NaSS: sodium p-styrenesulfonate; UM: 2-ureidoethyl methacrylate

Note: “N/A” indicates “not available” in the references

References:

1. Zeng, Z. F.; Yang, Y. Q.; Pang, X. W.; Jiang, B.; Gong, L. X.; Liu, Z.; Peng, L.; Li, S. N., Lignin Nanosphere‐Modified MXene Activated‐Rapid Gelation of Mechanically Robust, Environmental Adaptive, Highly Conductive Hydrogel for Wearable Sensors Application. *Adv. Funct. Mater.* **2024**, 2409855.

2. Ni, Q.-Y.; He, X.-F.; Zhou, J.-L.; Yang, Y.-Q.; Zeng, Z.-F.; Mao, P.-F.; Luo, Y.-H.; Xu, J.-M.; Jiang, B.; Wu, Q., Mechanical tough and stretchable quaternized cellulose nanofibrils/MXene conductive hydrogel for flexible strain sensor with multi-scale monitoring. *J. Mater. Sci. Technol.* **2024,** *191*, 181-191.

3. Luan, H.; Zhang, D.; Xu, Z.; Zhao, W.; Yang, C.; Chen, X., MXene-based composite double-network multifunctional hydrogels as highly sensitive strain sensors. *J. Mater. Chem. C* **2022,** *10* (19), 7604-7613.

4. Luo, Y.; Yang, Q.; Chen, M.; Long, K.; Su, C.; Li, J.; Huang, M.; Lu, A.; Guo, S., Stretchable, adhesive, conductive hydrogel initiated by liquid metal complex for multi-functional sensing. *Chem. Eng. J.* **2024,** *496*, 153674.

5. Chen, G.; Huang, J.; Gu, J.; Peng, S.; Xiang, X.; Chen, K.; Yang, X.; Guan, L.; Jiang, X.; Hou, L., Highly tough supramolecular double network hydrogel electrolytes for an artificial flexible and low-temperature tolerant sensor. *J. Mater. Chem. A* **2020,** *8* (14), 6776-6784.

6. Zhao, H.; Hao, S.; Fu, Q.; Zhang, X.; Meng, L.; Xu, F.; Yang, J., Ultrafast

fabrication of lignin-encapsulated silica nanoparticles reinforced conductive hydrogels with high elasticity and self-adhesion for strain sensors. *Chem. Mater.* **2022,** *34* (11), 5258-5272.

7. Wu, S.; Shao, Z.; Xie, H.; Xiang, T.; Zhou, S., Salt-mediated triple shape-memory ionic conductive polyampholyte hydrogel for wearable flexible electronics. *J. Mater. Chem. A* **2021,** *9* (2), 1048-1061.

8. Yang, W. J.; Zhang, R.; Guo, X.; Ma, R.; Liu, Z.; Wang, T.; Wang, L., Supramolecular polyelectrolyte hydrogel based on conjoined double-networks for multifunctional applications. *J. Mater. Chem. A* **2022,** *10* (44), 23649-23665.