Robust hydrogel sensor with high mechanical properties, conductivity, anti-swelling ability, water tolerance and biocompatibility

Airong Xu[a,*] Tiantian Sun,[a] Rukuan Liu[b,*] Liuzemu Li,[b] Yiming Gong,[a] and Zhihong Xiao

[a] *School of Chemistry and Chemical Engineering, Henan University of Science and Technology, Luoyang, Henan 471003, P. R. China*

[b] *State Key Laboratory of Utilization of Woody Oil Resource, Hunan Academy of Forestry, Changsha, Hunan 410004, P. R. China*

Table S1 Conductivity summary of the hydrogel in this work and PVA based hydrogels reported previously

Hydrogel	Conductivity (S m^{-1})	Ref.
PVA/SG	4.85	This work
PEDOT:PSS/PVA DN	0.1	1
PVA/SA	0.3-3.62	2
PVA/CNF@PDA/CNT	0.4	3
TOCNF/PANI/PVA/borax (CPPH)	0.6	4
PVA/HLC/TA/Borax (PHTB)	0.08	5
PVA-PAANa-PAH	0.3	6
PVA/CNC	0.46 – 1.02	7
PVA-PHMG-ADSP	0.125	8
AgNPs@CNTs/PVA/Glycerol/Borax	0.1667	9
Starch/PVA/ionic liquid/GO	1.98	10
PANI@(PVA–PAA)	2.02	11
nanocellulose/PVA/carbon dot	2.3×10^{-5}	12
PVA–NaCl–GL–AMY	3.5	13
PAAM/PNA/PVA/PANI	3.91	14
PVA+ P(SBMA-co-HEMA)	4.58	15
CNC-PEDOT:PSS/PVA	4.73	16
PVA/PAAS/Ca^{2+}	5.2	17
PVA-LMPs	5.4×10^{-4}	18
PVA/HEC	5.77	19
PVA/orthophosphoric acid	6.47×10^{-4}	20
PVA/polyaniline	9.5. 19.0, and 20.5	21
PVA-PEG-SA-180	15	22
CMC/PVA	< 5.4	23
PVA-PANa	1.65	24
PVA-PA	1.34 kΩ·cm	25
PMZn-GL	0.056	26
PVA/PAMAA	3-15	27
HPC/PVA	3.4	28
LiBr@PVAM	0.62-0.95	29
PAM/PVA/CNF	0.0297	30

Table S2 Summary of the stretching stress, stretching strain, elastic modulus and toughness of the hydrogel in this work and PVA based hydrogels reported previously

Hydrogel	Stretching stress (MPa)	Stretching strain (%)	Elastic modulus (MPa)	Toughness (MJ m^{-3})	Ref.
PVA/SG	1.23	387	0.315	2.298	This work
PVA/SA	0.56-1.32	280-400	0.34-0.59	0.64-3.14	2
PVA/CNF@PDA/CNT	0.79	335-471	0.02023-0.08293	1.26	3
TOCNF/PANI/PVA/borax (CPPH)	0.01564	1530	a	a	4
PVA-PAANa-PAH	0.86	768	2.25	2.25	6
PVA/CNC	0.75	410.47	a	a	7
Starch/PVA/ionic liquid/GO	0.64	657.5	~ 0.275	< 2.25	10
PANI@(PVA–PAA)	0.09051	a	0.02373	a	11
nanocellulose/PVA/carbon dot	2.98	a	a	a	12
PVA+ P(SBMA-co-HEMA)	< 0.4	< 350	< 0.125	0.518	15
PVA/PAAS/Ca^{2+}	0.75	216	a	a	17
PVA/polyaniline	1.46	416	4.27	3.28	21
PVA-PEG-SA-180	16.1	204.93	a	a	22
PVA-PANa	7.0	600	a	a	24
PVA-PA	0.5-1.5	500-1100	a	a	25
HA-PVA	23.5	2900	a	210	31
PVA from salting-out	15	2100	2.5	20	32
PVA/PAA from cold-drawing	140	a	100	117	33
PVA from cold-drawing	5	a	0.5	a	34
PMZn-GL	0.875	247	$< 1 \times 10^{-9}$	0.082	26
PVA/PAMAA	0.4363	608	0.1349	0.0047	27
HPC/PVA	1.3	975	0.9	5.85	28
PAM/PVA/CNF	9.43	445.5	a	15.12	30
PAM-PVA	0.1838	488	0.087	a	35
PVA-gelatin-Fe^{3+}	2.5	255	0.0252	7.22	36
PVA/PAA-PEDOT:PSS-TA	0.202-0.24	443-640	a	0.0494-0.01231	37
PVA−TA300	9.5	1000	~1.7	a	38

a Not investigated, and/or no data reported.

Table S3 Summary of sensitivity (gauge factor) of the hydrogel in this work and PVA based hydrogels reported previously

Hydrogel	Sensitivity (gauge factor)	Ref.
PVA/SG	0.91, 1.76	This work
PVA/CNF@PDA/CNT	2.29	3
PVA-PAANa-PAH	1.20, 1.64	6
Starch/PVA/ionic liquid/GO	6.04	10
PVA+ P(SBMA-co-HEMA)	1.434, 2.448, and 3.356	15
PVA/PAAS/Ca^{2+}	1.58	17
PVA-PANa	0.45	24
PMZn-GL	3.42, 4.77, 5.82	26
PAM/PVA/CNF	1.47, 2.72, 4.39,	30
PVA/PAA-PEDOT:PSS-TA	1.29, 2.34	37

Table S4 Summary of fatigue resistance of the hydrogel in this work and PVA based hydrogels reported previously

Hydrogel	Fatigue resistance	Ref.
PVA/SG	Yes	This work
PVA/CNF@PDA/CNT	Yes	4
Starch/PVA/ionic liquid/GO	Yes	10
PANI@(PVA–PAA)	Yes	11
PVA/PAAS/Ca^{2+}	Yes	17
PVA/polyaniline	Yes	21
PVA-PANa	Yes	24
HA-PVA	Yes	31
PVA/PAMAA	Yes	27
PVA-gelatin-Fe3+	Yes	36
PVA/PAA-PEDOT:PSS-TA	Yes	37

Table S5 Summary of swelling resistance and water resistance of the hydrogel in this work and PVA based hydrogels reported previously

Hydrogel	Swelling resistance in water	Water resistance	Ref.
PVA/SG	Yes	Still remain good enough mechanical properties after six months in water	This work
PVA/SA	Swelling	b	2
PVA-PAANa-PAH	Swelling	Apparently decreased in stress and strain	6
PVA+ P(SBMA-co-HEMA)	Yes	Decrease in stress and strain	15
PVA/PAAS/Ca^{2+}	Yes	Still remain good enough strain,but apparently decreased stress after 24 h in water	17
PVA-PEG-SA-180	Swelling	b	22
PVA-PANa	a	b	24
PVA/PAA	Swelling	Decrease in stress and strain	34
PVA/PAMAA	Swelling	b	27
PVA-gelatin-Fe^{3+}	Swelling	b	36

a Antisweling in PBS buffer and 0.9% NaCl solution. b Not investigated, and/or no data reported.

References

[1] G. Li, K. Huang, J. Deng, M. Guo, M. Cai, Y. Zhang, C. F. Guo, *Adv. Mater.* 2022, **34**, 2200261.

[2] X. Jiang, N. Xiang, H. Zhang, Y. Sun, Z. Lin, L. Hou, *Carbohyd. Polym.* 2018, **186**, 377.

[3] R. Zhang, A. Yang, Y. Yang, Y. Zhu, Y. Song, Y. Li, J. Li, *Int. J. Biol. Macromol.* 2023, **245**, 125469.

[4] Y. Qin, J. Mo, Y. Liu, S. Zhang, J. Wang, Q. Fu, S. Wang, S. Nie, *Adv. Funct. Mater.* 2022, **32**, 2201846.

[5] H. Lei, D. Fan, *Chem. Eng. J.* 2021, **421**, 129578.

[6] W. J. Yang, R. Zhang, X. Guo, R. Ma, Z. Liu, T. Wang, L. Wang, *J. Mater. Chem. A*

2022, **10**, 23649.

[7] X. Huang, X. Ao, L. Yang, J. Ye, C. Wang, *RSC Adv.* 2023, **13**, 527.

[8] Y. Gao, Y. Gao, Z. Zhang, F. Jia, G. Gao, *ACS Appl. Mater. Inter.* 2022, **14**, 51420.

[9] X. Yu, W. Qin, X. Li, Y. Wang, C. Gu, J. Chen, S. Yin, *J. Mater. Chem. A* 2022, **10**, 15000.

[10] X. Li, S. Zhang, X. Li, L. Lu, B. Cui, C. Yuan, L. Guo, B. Yu, Q. Chai, *Carbohyd. Polym.* 2023, **320**, 121262.

[11] X. Dong, Y. Ge, K. Li, X. Li, Y. Liu, D. Xu, S. Wang, X. Gu, *Soft Matter* 2022, **18**, 9231.

[12] Z. Wang, F. Cheng, H. Cai, X. Li, J. Sun, Y. Wu, N. Wang, Y. Zhu, *Carbohyd. Polym.* 2021, **259**, 117753.

[13] Y. Gao, Y. Gao, Z. Zhang, Y. Wang, X. Ren, F. Jia, G. Gao, *J. Mater. Chem. C* 2022, **10**, 12873.

[14] C. Qian, Y. Li, C. Chen, L. Han, Q. Han, L. Liu, Z. Lu, *Chem. Eng. J.* 2023, **454**, 140263.

[15] J. Ren, Y. Liu, Z. Wang, S. Chen, Y. Ma, H. Wei, S. Lü, *Adv. Funct. Mater.* 2021, **32**, 2107404.

[16] X. Chai, J. Tang, Y. Li, Y. Cao, X. Chen, T. Chen, Z. Zhang, *ACS Appl. Mater. Inter.* 2023, **15**, 18262.

[17] R. Liu, K. Chen, H. Liu, Y. Liu, R. Cong, J. Guo, Y. Tian, *ACS Appl. Mater. Inter.* 2022, **14**, 51341.

[18] M. Liao, H. Liao, J. Ye, P. Wan, L. Zhang, *ACS Appl. Mater. Inter.* 2019, **11**, 47358.

[19] X. Wang, X. Wang, M. Pi, R. Ran, *Chem. Eng. J.* 2022, **428**, 131172.

[20] M. Z. Kufian, S. R. Majid, A. K. Arof, *Ionics* 2007, **13**, 231.

[21] L. Li, Y. Zhang, H. Lu, Y. Wang, J. Xu, J. Zhu, C. Zhang, T. Liu, *Nature Commun.* 2020, **11**, 62.

[22] Y. Xu, M. Pei, J. Du, R. Yang, Y. Pan, D. Zhang, S. Qin, *New J. Chem.* 2023, **47**, 13721.

[23] H. Patnam, S. A. Graham, P. Manchi, M. V. Paranjape, J. S. Yu, *ACS Appl. Mater. Inter.* 2023, **15**, 16768.

[24] S. Zhang, Y. L. Li, H. J. Zhang, G. J. Wang, H. Wei, X. Y. Zhang, N. Ma, *ACS Mater.*

Lett. 2021, **3**, 807-814.

[25] S. Zhang, Y. H. Zhang, B. Li, P. Zhang, L. Kan, G. J. Wang, H. Wei, X. Y. Zhang, N. Ma, *ACS Appl. Mater. Interfaces* 2019, **11**, 32441-32448.

[26] Y. B. Feng, H. Liu, W. H. Zhu, L. Guan, X. T. Yang, A. V. Zvyagin, Y. Zhao, C. Shen, B. Yang, Q. Lin, *Adv. Funct. Mater.* 2021, **31**, 2105264.

[27] J. R. Huang, S. J. Han, J. D. Zhu, Q. R. Wu, H. J. Chen, A. B. Chen, J. Y. Zhang, B. Huang, X. X. Yang, L. H. Guan, *Adv. Funct. Mater.* 2022, **32**, 2205708.

[28] Y. Zhou, C. J. Wan, Y. S. Yang, H. Yang, S. C. Wang, Z. D. Dai, K. J. Ji, H. Jiang, X. D. Chen, Y. Long, *Adv. Funct. Mater.* 2018, **29**, 1806220.

[29] A. A. M. Ismail, L. G. Ghanem, A. A. Akar, G. E. Khedr, M. Ramadan, B. S. Shaheen, N. K. Allam, *J. Mater. Chem. A* 2023, **11**, 16009–16018.

[30] J. M. Wu, Q. Ma, Q. K. Pang, S. S. Hu, Z. H. Wan, X. F. Peng, X. Cheng, L. H. Geng, *Carbohydr. Polym.* 2023, **321**, 121282.

[31] M. T. Hua, S. W. Wu, Y. F. Ma, Y. S. Zhao, Z. L. Chen, I. Frenkel, J. Strzalka, H. Zhou, X. Y. Zhu, X. M. He, *Nature* 2021, **590**, 594 - 599.

[32] S. W. Wu, M. T. Hua, Y. Alsaid, Y. J. Du, Y. F. Ma, Y. S. Zhao, C.-Y. Lo, C. R. Wang, D. Wu, B. W. Yao, J. Strzalka, H. Zhou, X. Y. Zhu, X. M. He, *Adv. Mater.* 2021, **33**, 2007829.

[33] T. Q. Liu, C. Jiao, X. Peng, Y. N. Chen, Y. Y. Chen, C. C. He, R. G. Liu, H. L. Wang, *J. Mater. Chem. B* 2018, **6**, 8105-8114.

[34] Y. Y. Chen, C. Jiao, X. Peng, T. Q. Liu, Y. Q. Shi, M. Liang, H. L. Wang, *J. Mater. Chem. B* 2019, **00**, 1-3.

[35] 35G. Ge, Y. Z. Zhang, J. J. Shao, W. J. Wang, W. L. Si, W. Huang, X. C. Dong, *Adv. Funct. Mater.* 2018, **28**, 1802576.

[36] 36S. Sun, Y. Z. Xu, X. Maimaitiyiming, *Int. J. Biol. Macromol.* 2023, **249**, 125978.

[37] 37Z. X. Zhou, W. Z. Yuan, X. Y. Xie, *Mater. Chem. Front.* 2022, **6**, 3359–3368.

[38] 38H. L. Fan, J. H. Wang, Z. X. Jin, *Macromolecules* 2018, **51**, 1696-1705