Electronic Supporting Information

Flexible hydrogel tactile sensor with low compressive modulus and dynamic piezoresistive response regulated by lignocellulose/graphene aerogel

Hang Zhou[#a], Li Zheng[#a], Qingyu Meng[a], Ruixin Tang[a], Zhaosong Wang[a], Baokang Dang[a], Xiaoping Shen[a*], Qingfeng Sun[a*]

[a]School of Engineering, Zhejiang A&F University, Hangzhou 311300, People's Republic of China

*Corresponding authors: Xiaoping Shen, E-mail: xpshen@zafu.edu.cn; Qingfeng Sun, E-mail: qfsun@zafu.edu.cn

[#] H. Zhou and L. Zheng contributed equally to this work.

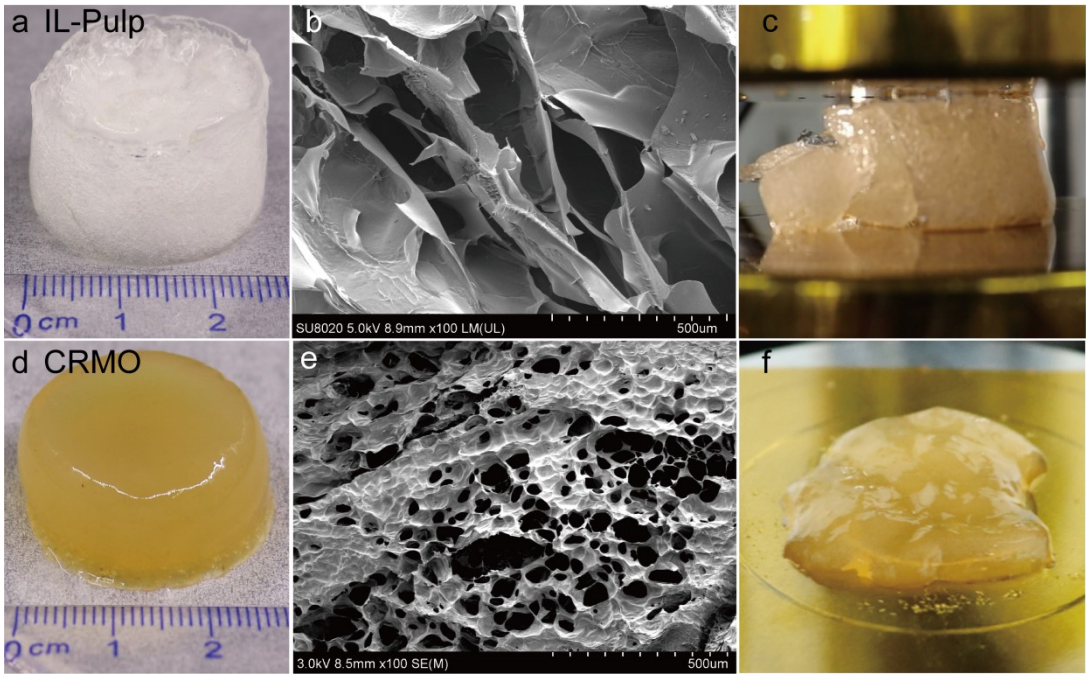

Fig. S1 The SEM images and the appearance pictures of the hydrogel from ionic liquid (IL)-regenerated pulp (IL-Pulp) and the original CRM hydrogel (CRMO) before and after compression. IL-Pulp was prepared by dissolving pulp in [Bmim]Cl and regenerating in H_2O.

Table S1 Determination of component contents (%) of the lignocellulosic hydrogels.

Component	Wood	CRM	CRMO	CRMRH	IL-pulp
Cellulose	49.2	60.8	82.8	83.1	100
hemicellulose	30.2	27.0	8.3	8.1	0
lignin	18.3	12.2	8.9	8.8	0

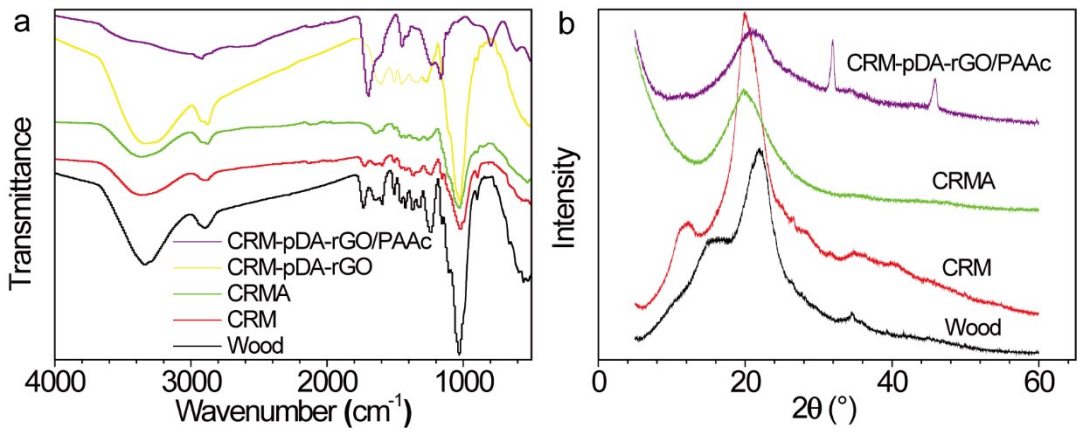

Fig. S2 (a) FT-IR and (b) XRD of the lignocellulosic hydrogels during the whole processes.

Table S2 Gradient sensitivity values of the ionic hydrogel sensors.

Samples	S1 (MPa^{-1})/Range (MPa)	S2 (MPa^{-1})/Range (MPa)
PAAc	1.26/0.047	0.003/0.21
CRM/PAAc	4.18/0.034	0.003/0.57
CRM-pDA-rGO/PAAc	9.71/0.022	0.012/0.81

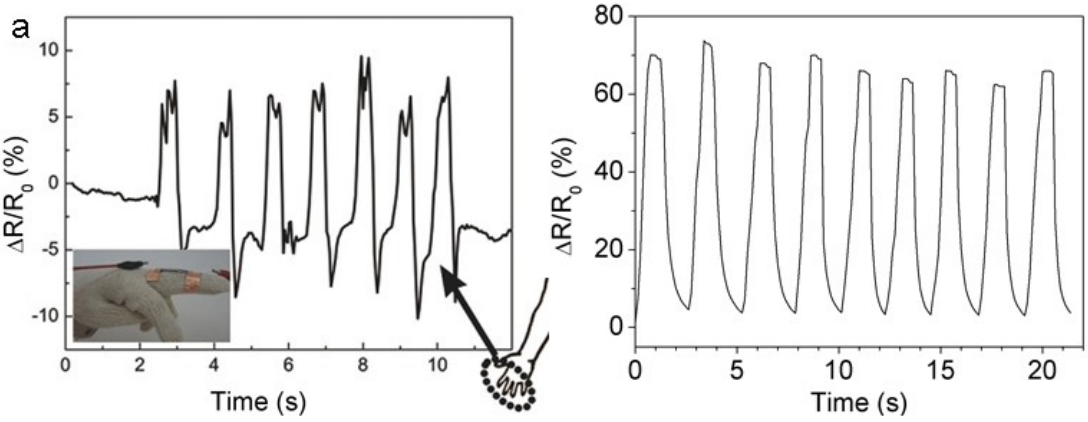

Fig. S3 Relative resistance changes versus time for the bending and release of the index finger using (a)

nanocarbon-contained hydrogel sensors as reported in literature,[1] or (b) using our hydrogel sensor.

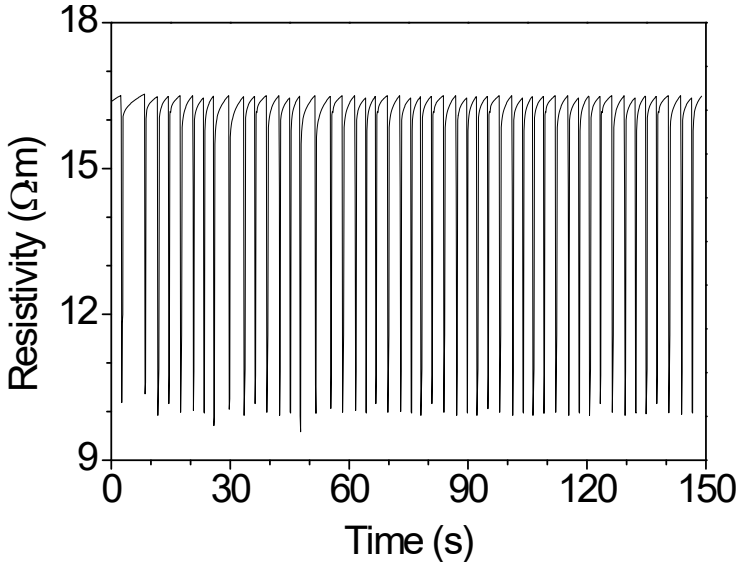

Fig. S4 Sensing durability of the hydrogel sensor under *ca.* 0.2 MPa stress over 50 cycles.

Table S3 Comparison of sensitivity and stress ranges of various hydrogel sensors.

Reference	Type of sensor	Materials	Stress range	Sensitivity
[2]	Capacitive	PAAc/alginate/ACC hydrogel	1.0 KPa	0.17 KPa^{-1}
[3]	Capacitive	PAAm-LiCl hydrogel	26 KPa	0.006 KPa^{-1}
[4]	Capacitive	PAAm/NaCl hydrogel	40 KPa	0.009 KPa^{-1}
[5]	Capacitive	Gelatin DES gel	160 KPa	0.013 KPa^{-1}
[6]	Capacitive	AgNWs/Ecoflex	1.2 MPa	1.62 MPa^{-1} (<0.4 MPa) 0.57 MPa^{-1} (<1.2 MPa)
[7]	Piezoresistive	cellulose/graphene composites	35 MPa	0.0013 MPa^{-1}
[8]	Piezoresistive	PVA/PAAc/F-MWCNT/PEDOT	25 KPa	0.011 KPa^{-1}
[9]	Piezoresistive	Carbon aerogel (TMCA)	15 KPa	0.00131 KPa^{-1}
[10]	Piezoresistive	CSM/PAAm/PANI hydrogel	6 MPa	0.05 KPa^{-1} (<10 kPa) 10^{-5} KPa^{-1} (>0.5 MPa)
[11]	Piezoresistive	PEI/CNT	40 MPa	0.05 MPa^{-1}

[12]	Piezoresistive	Pt-rGOH aerogel	1.8 MPa	0.3 MPa^{-1}
[13]	Piezoresistive	PAniNR@PAN	0.3 MPa	0.95 MPa^{-1}
Our work	Piezoresistive	CRM-pDA-rGO/PAAc hydrogel	0.8 MPa	9.34 MPa^{-1} (<25 KPa) 9.71 MPa^{-1} (<0.022 MPa) 0.012 MPa^{-1} (<1.22 MPa)

Abbreviations: polyacrylic acid (PAAc), calcium carbonate (ACC), polyacrylamide (PAAm), deep eutectic solvent (DES), nanowires (NWs), polyvinyl alcohol (PVA), surfactantfunctionalized multi-walled carbon nanotube (F-MWCNT), poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS), TEMPO-oxide cellulose nanofibers (TOCN)-coated-melamine foams (MF) carbon aerogels (TMCA), chitosan microspheres (CSM), polyaniline (PANI), polyethyleneimine (PEI), carbon nanotube (CNT), Pt/reduced graphene oxide hydrogel (Pt-rGOH), polyaniline nanorod on polyacrylonitrile (PAN) nanofiber substrate (PAniNR@PAN)

References

1. G. Cai, J. Wang, K. Qian, J. Chen, S. Li and P. S. Lee, *Adv. Sci.*, 2017, 4, 1600190.
2. Z. Lei, Q. Wang, S. Sun, W. Zhu and P. Wu, *Adv. Mater.*, 2017, 29, 1700321.
3. C. Larson, B. Peele, S. Li, S. Robinson, M. Totaro, L. Beccai, B. Mazzolai and R. Shepherd, *Science*, 2016, 351, 1071-1074.
4. J. Y. Sun, C. Keplinger, G. M. Whitesides and Z. Suo, *Adv. Mater.*, 2014, 26, 7608-7614.
5. H. Qin, R. E. Owyeung, S. R. Sonkusale and M. J. Panzer, *J. Mater. Chem. C.*, 2019, 7, 601-608.
6. S. Yao and Y. Zhu, *Nanoscale*, 2014, 6, 2345-2352.
7. Y. Chen, P. Pötschke, J. Pionteck, B. Voit and H. Qi, *J. Mater. Chem. A*, 2018, 6, 7777-7785.

8. G. Ge, W. Yuan, W. Zhao, Y. Lu, Y. Zhang, W. Wang, P. Chen, W. Huang, W. Si and X. Dong, *J. Mater. Chem. A*, 2019, 7, 5949-5956.

9. M. Wang, Y. Chen, Y. Qin, T. Wang, J. Yang and F. Xu, *ACS Sustain. Chem. Eng.*, 2019, *7*(15), 12726-12733.

10. J. Duan, X. Liang, J. Guo, K. Zhu and L. Zhang, *Adv. Mater.*, 2016, *28*(36), 8037-8044.

11. S. M. Doshi and E. T. Thostenson, *ACS Sens.*, 2018, 3, 1276-1282.

12. S.-H. Hwang, Y.-B. Park, S.-H. Hur and H. G. Chae, *ACS Appl. Nano Mater.*, 2018, 1, 2836-2843.

13. H. H. Shi, N. Khalili, T. Morrison and H. E. Naguib, *ACS Appl. Mater. Interfaces*, 2018, 10, 19037-19046.