Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.


# Transparent, ultrastretchable and fully recyclable gelatin organohydrogel based electronic sensor with broad operating temperature

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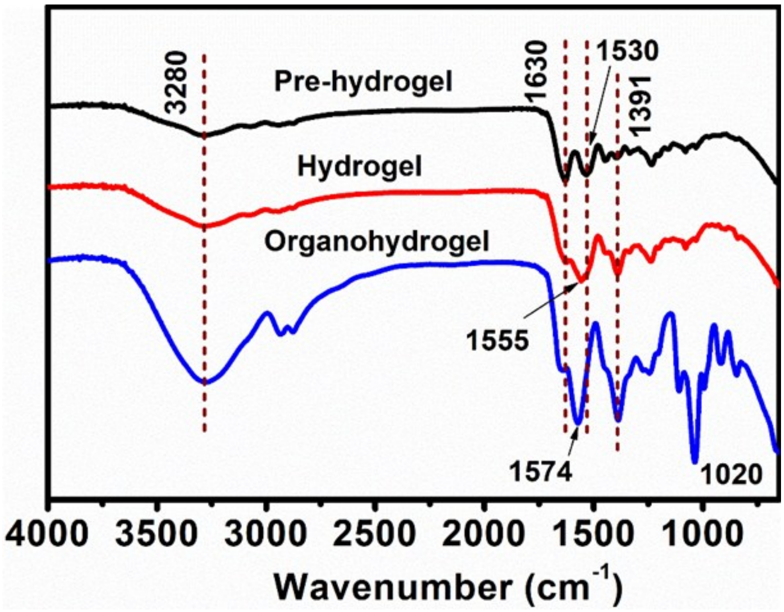


**Fig. S1** FTIR spectra of gelatin pre-hydrogel, hydrogel and organohydrogel.

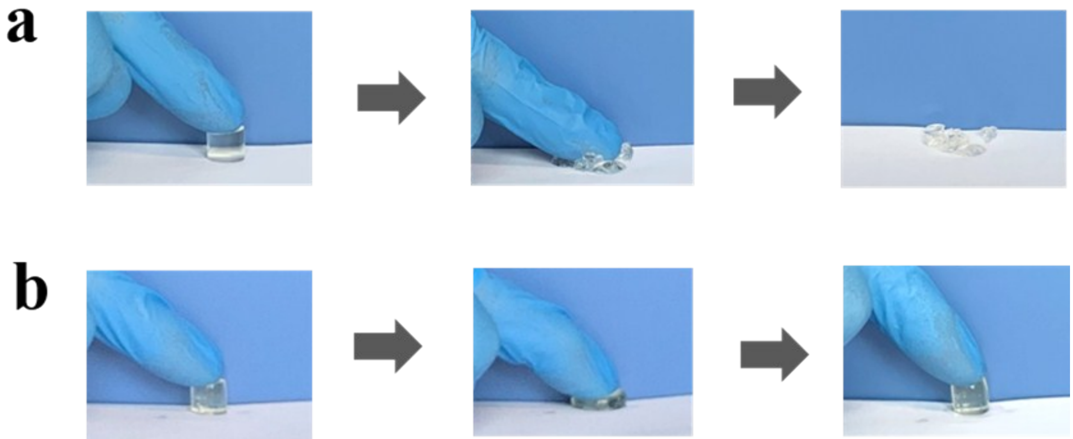


**Fig. S2** Photographs of compression performances of gelatin (a) pre-hydrogel and (b) organohydrogel

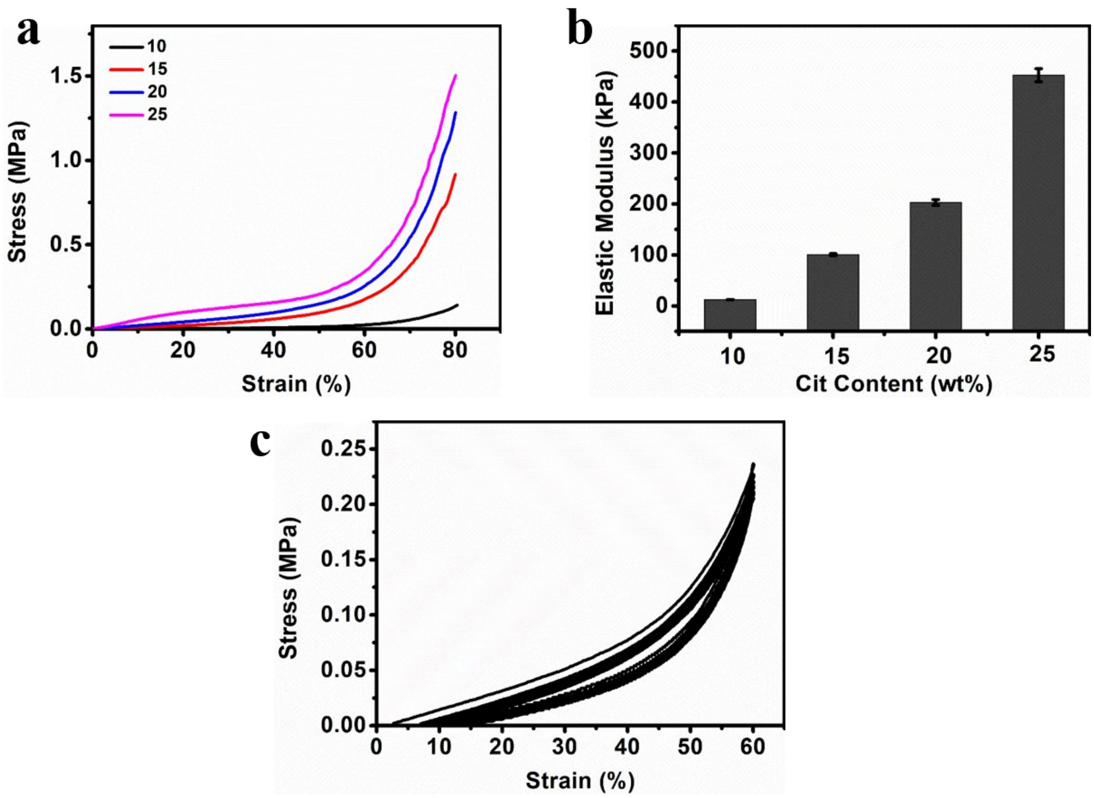


**Fig. S3** (a) The compressive stress-strain curves and (b) the corresponding elastic modulus of the gelatin organohydrogels soaked in different Na3Cit concentration solutions. (c) Ten successive cyclic compressive tests of the gelatin organohydrogel.

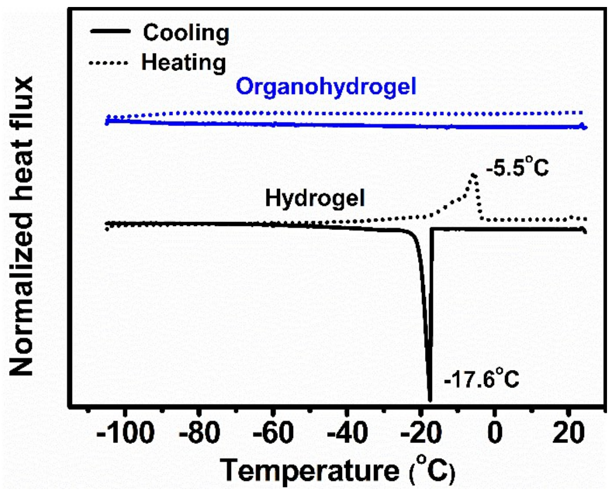


**Fig. S4** DSC thermograms of gelatin hydrogel and organohydrogel.

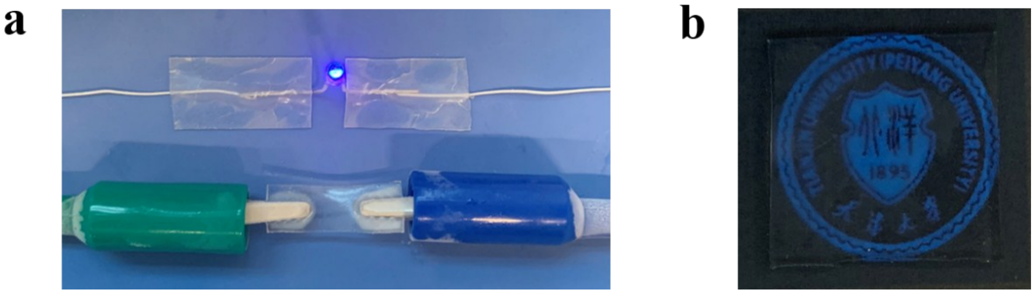


**Fig. S5** Photographs showing (a) the conductivity and (b) transparency of the organohydrogel at -60 °C.

**Table S1.** Comparison of reported nature-driving hydrogels with this work.

| Composition | Tensile strength (MPa) | Tensile strain (%) | Elastic modulus (kPa) | Conductivity (S/m) | Transparency (%) | Ref. |
|---|---|---|---|---|---|---|
| **Gelatin/$Na_3$Cit** | **1.91** | **542** | **91.0** | **0.47** | **96** | **This work** |
| MeTro/GO | 0.02 | 203 | 19.3 | / | / | 1 |
| allyl cellulose | ~0.05 | 126 | 21 | 0.016 | 89 | 2 |
| RLPs/rGO | 0.088 | 326 | 44 | 0.92 | / | 3 |
| Alg/Ca | 2.43 | 400 | 1090 | / | / | 4 |
| EW/DMEM | 0.014 | 87 | 16.9 | / | / | 5 |
| Cellulose/$ZnCl_2$ | 0.28 | 120 | 500 | 7.49 | 84 | 6 |
| RSF/HPMC | 0.23 | 129 | 750 | / | / | 7 |
| GelMA/TA | ~0.15 | 225 | 80 | / | / | 8 |
| Cellulose/$BzMe_3NOH$ | 2.0 | 219 | / | 2.37 | / | 9 |
| Cellulose/ECH | 2.7 | 81 | 2000 | / | / | 10 |
| SF | 0.56 | 127 | 2600 | / | / | 11 |
| Chitosan /ECH | 0.37 | 117 | 200 | / | / | 12 |
| RSF/SDS | 0.74 | 134 | 3280 | / | / | 13 |
| Gelatin/DES | 0.067 | 320 | / | 0.25 | 80~90 | 14 |

Note: “/” indicates “not shown” in the references.

MeTro: methacryloyl-substituted recombinant human tropoelastin, RLPs: reslin-like proteins, Alg: alginate, EW: egg white, DMEM: Dulbecco's Modified Eagle Medium, RSF: regenerated silk fibroin, HPMC: hydroxypropyl methyl cellulose, GelMA: gelatin methacrylate, TA: tannic acid, $BzMe_3NOH$: benzyltrimethyl ammonium hydroxide, ECH: epichlorohydrin, SF: silk fibroin, DES: deep eutectic solvent.

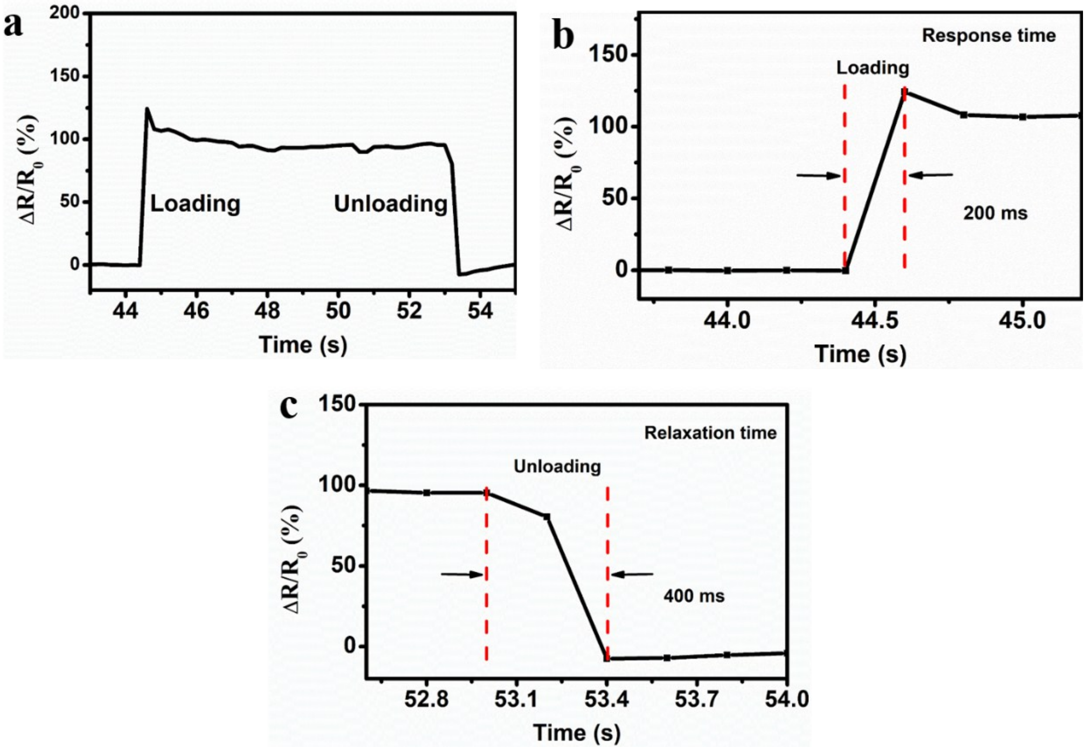


**Fig. S6** Sensitivity of the organohydrogel-based strain sensor. (a) $\Delta R/R_0$ response in instantaneous loading-unloading process at 1000% strain. (b) The response time for loading and (c) the relaxation time for unloading of the strain sensor.

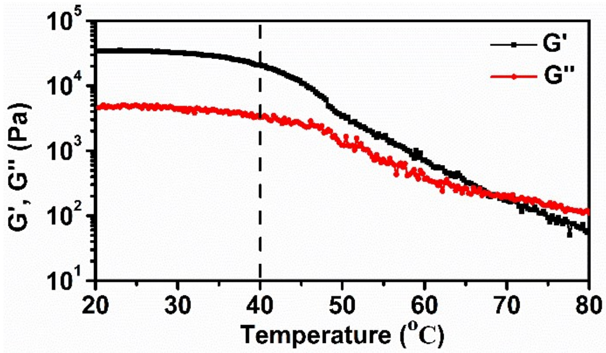


**Fig. S7** Storage modulus (G') and loss modulus (G") of gelatin organohydrogel on a temperature sweep at a constant shear strain of 0.1 % and angular frequency of 1 Hz.

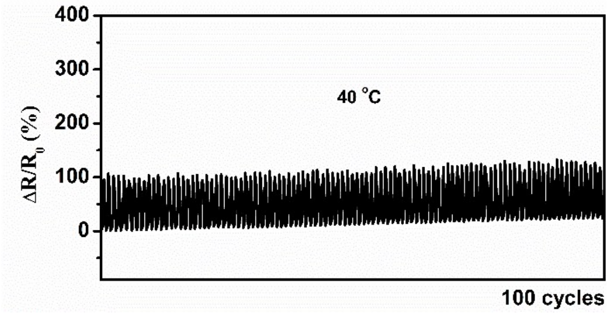


**Fig. S8** Durability test of gelatin organohydrogel-based electronic sensor under repeated strains of 100% for 100 cycles at 40 °C.

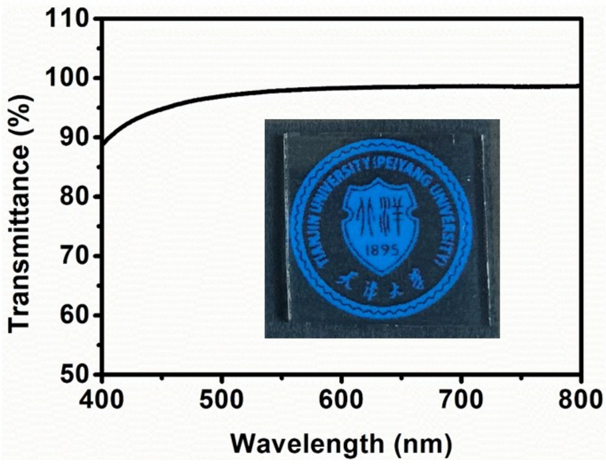


**Fig. S9** Transparency of the recycled organohydrogel.

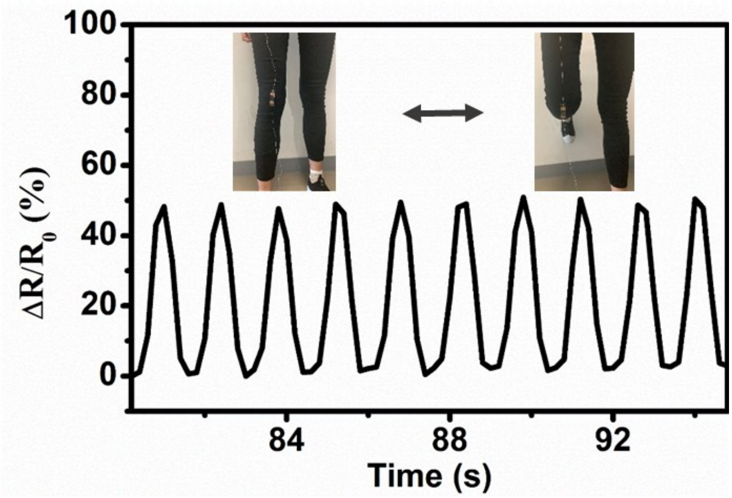


**Fig. S10** Wearable electronic sensor for detecting knee bending.

**Table S2.** Comparison of reported flexible electronic sensors with this work in functionalities.

| Composition | Renewable biomass | Stretchability (%) | Transparency (%) | Low-temperature operating | Recyclability | Ref. |
|---|---|---|---|---|---|---|
| **Gelatin/$Na_3Cit$** | **Yes** | **300** | **96** | **Yes (-30 °C)** | **Yes (Fully)** | **This work** |
| allyl cellulose | Yes | 90 | 89 | Yes (-20 °C) | - | 2 |
| Cellulose/$BzMe_3NOH$ | Yes | 219 | 90 | Yes (-27.8 °C) | - | 9 |
| $TiO_2$-CD/CS/PVA | - | 278 | - | - | - | 15 |
| PANI/PSS−UPy | - | 300 | - | - | - | 16 |
| SA/NaCl/PAM | - | 1800 | 99.6 | - | - | 17 |
| HP(AAm/AA)-CS-$Fe^{3+}$ | - | 500 | - | - | - | 18 |
| PVA/TA@talc | - | 100 | Yes | Yes (-30 °C) | - | 19 |
| PAAm/PVA/MXene | - | 350 | - | Yes (-40 °C) | - | 20 |
| PAAm/PVA/CNT/ PEDOT: PSS | - | 550 | - | Yes (-25 °C) | - | 21 |
| PVA/PAANa-$Tb^{3+}$ | - | ~350 | - | - | Yes | 22 |
| PAA-alginate-EGaIn | - | 300 | - | - | Yes | 23 |
| PAM/Au@PDA | - | - | - | Yes (-15 °C) | - | 24 |
| HPC/PVA | - | 400 | - | - | - | 25 |
| PEDOT:SL-PAA | - | 100 | - | Yes (-15 °C) | - | 26 |
| PVA/PMA | - | 500 | 90 | - | - | 27 |
| PAAm/PEO/LiCl | - | 880 | - | - | - | 28 |
| PANI/P(AAm-co-HEMA) | - | 330 | - | - | - | 29 |
| PVA/GE/GL | - | 700 | - | Yes (-20 °C) | - | 30 |
| PAM/Carrageenan | - | 400 | - | Yes (-18°C) | - | 31 |

Note: “-” indicates “not available” in the references.

$BzMe_3NOH$: benzyltrimethyl ammonium hydroxide, CS: chitosan, SA: sodium alginate, EGaIn: super conductive liquid metal, PDA: polydopamine, HPC: hydroxypropyl cellulose, SL: sulfonated lignin.

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