Electronic Upcycling of Corn Straw: Biobased Elastic Conductive Cross-linked Network Eco-friendly Hydrogels with multifunction for Flexible Wearable Sensors

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Supporting information

S1. Fabrication of different experimental samples

In order to intuitively explore the interactions between the elastic materials in the different experimental groups, we used only the base material (PVA, CSP and cellulose) in the preparation of the samples (PVA-Bond, PVA/Cellulose-Bond and PVA/CSP-Bond). After heating in a water bath at 80 °C and thorough mixing for 2 h, they were dried to form a film for further material characterization experiments.

In the fabrication of PVA -SYS, PVA/Cellulose-SYS and PVA/CSP-SYS, only different elastic substrates were used, and the rest of the process remained the consistent as described in the main text. Since the cellulose content in CSP is 70%, its amount was controlled at 0.7 g in the experimental group containing cellulose.

Table 1. Experimental sample composition summary

Sample	*PVA (g)*	*Cellulose (g)*	*CSP (g)*	*H_3PO_4 (ml)*	*$C_3H_8O_3$ (ml)*	*A-WMCNTs (ml)*	*5%wt $NaHCO_3$(ml)*	*Water (ml)*
PVA-Bond	1	0	0	1.5	1.5	0	0	20
PVA/Cellulose-Bond	1	0.7	0	1.5	1.5	0	0	20
PVA/CSP-Bond	1	0	1	1.5	1.5	0	0	20
PVA -SYS	1	0	0	1.5	1.5	1	10	20
PVA/Cellulose-SYS	1	0.7	0	1.5	1.5	1	10	20
PVA/CSP-SYS	1	0	1	1.5	1.5	1	10	20

When fabricating the conductive gel systems containing elastic base materials with different mass ratios, variations can be achieved only by adjusting the content of PVA and CSP, and remaining preparation processes and reagent use methods are consistent.

Notably, during the fabrication of samples with different ratios, we observed that when the percentage of CSP was too high (PVA:CSP < 0.6), the hydrogels prepared by the freeze-cycling process lacked elasticity and showed plastic characteristics, whereas a hydrogel system could not be formed when CSP was used completely.·In addition, we also found that the mechanical properties of the gels decreased with the increase of PVA proportion. When PVA is completely used to form a hydrogel, even small external stresses resulted in their destruction.

Table 2. CSP-MCG with elastic substrates of different mass ratios

The content ratio of PVA and CSP in gel system	*PVA(g)*	*CSP(g)*
The CSP content in the gel system is 100%	0	0.294
PVA:CSP=0.3	0.5	1.666
PVA:CSP=0.5	0.5	1.000
PVA:CSP=0.6	0.5	0.715
PVA:CSP=0.8	0.5	0.625
PVA:CSP=1.0	0.5	0.500
PVA:CSP=1.2	0.5	0.416
PVA:CSP=1.5	0.5	0.333
PVA:CSP=1.7	0.5	0.294
PVA:CSP=1.9	0.5	0.263
PVA:CSP=3	0.5	0.167
The PVA content in the gel system is 100%	0.5	0

S2. Mechanical flexibility of CSP-MCG

As shown in FIG. S1, the CSP-MCG can withstand a range of externally imposed deformations, such as knotting, twisting, and poking. It can even fold completely in half several times to lift a two-liter bottled water. Even in tensile tests, the maximum tensile rate can reach almost 340 %.

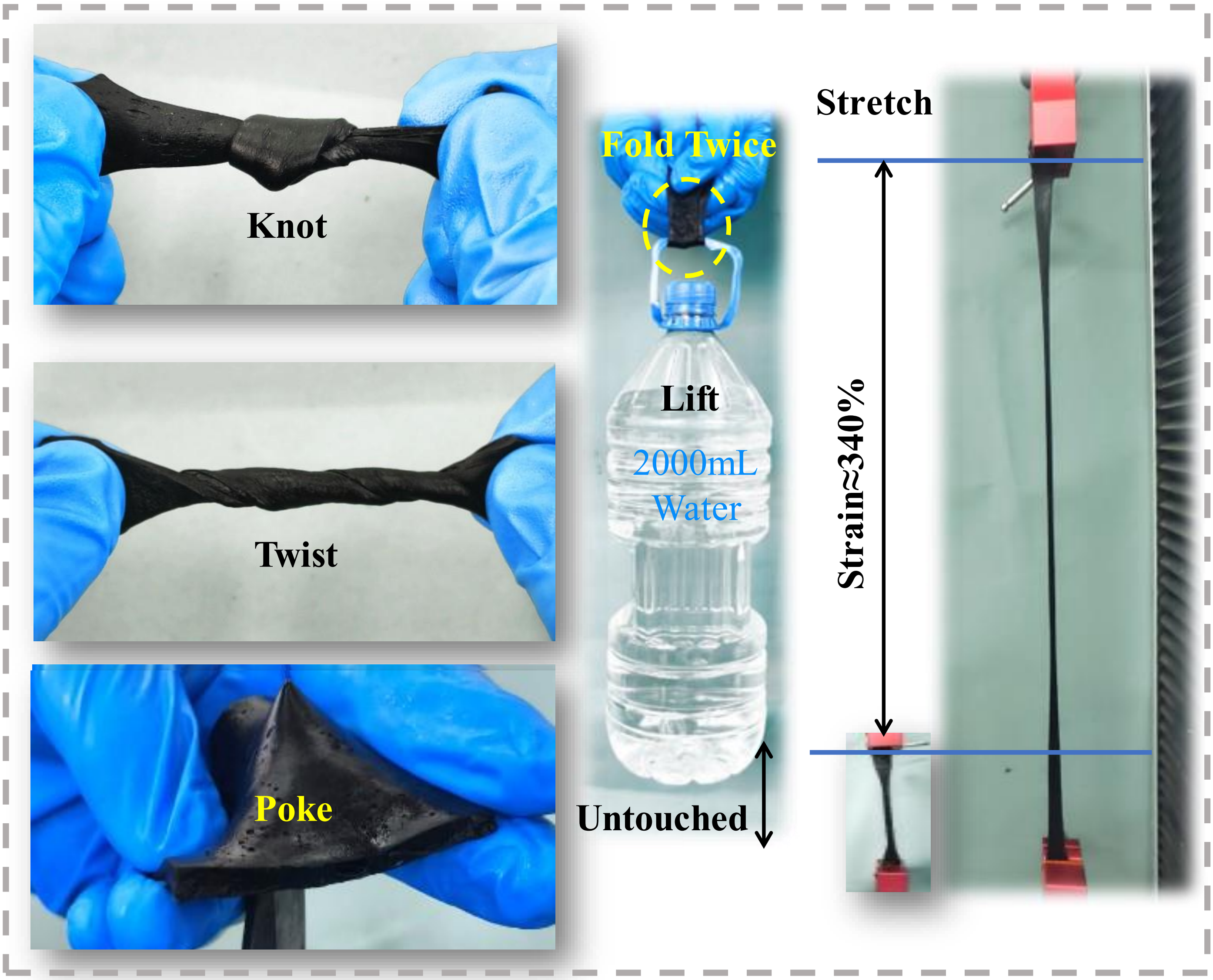

Figure S1. Photos of CSP-MCG knotting, twisting, poking, lifting and extreme tensile tests.

S3. Self-adhesive properties of CSP-MCG

As CSP-MCG is rich in hydroxyl and carboxyl groups, it shows good adhesive properties and can be bonded to the surface of different materials more easily. These include: glass sheets, plastic sheets, copper sheets, wood blocks, carbon fiber boards, aluminum sheets, rubber mold, foams, finger surfaces. As shown in the figure, when the gel adheres to the surface of a material, it provides a surface adhesion force sufficient to lift it against gravity.

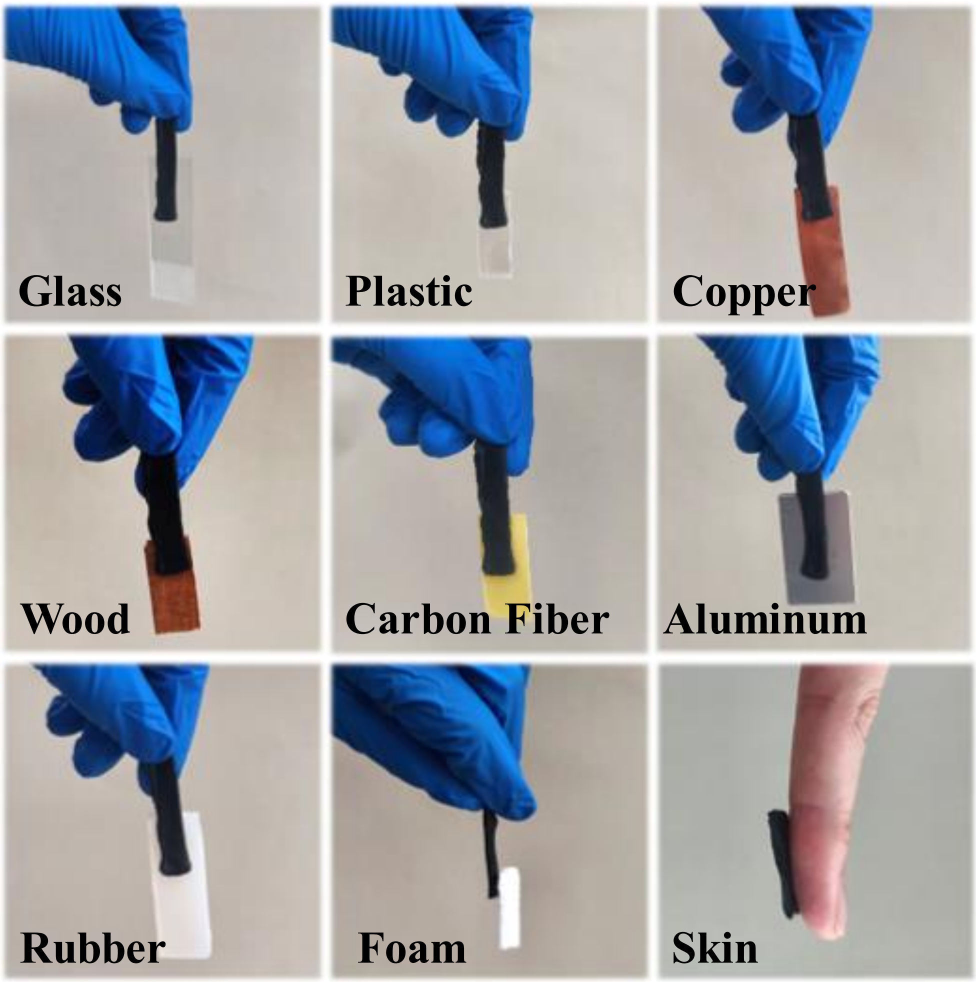

Figure S2. Photos of CSP-MCG adhesion to different substrates.

S4. Shear stress resulting from interfacial stripping off CSP-MCG adhering to different substrates

The self-adhesive property of hydrogels is a characteristic that is beneficial as a wearable sensing gel. The magnitude of surface peeling shear stresses for adherence to different materials is shown in the radial histogram below. As can be seen from the results in the figure, CSP-MCG is not only biocompatible but also has good adhesion when it is adhered to the human surface.

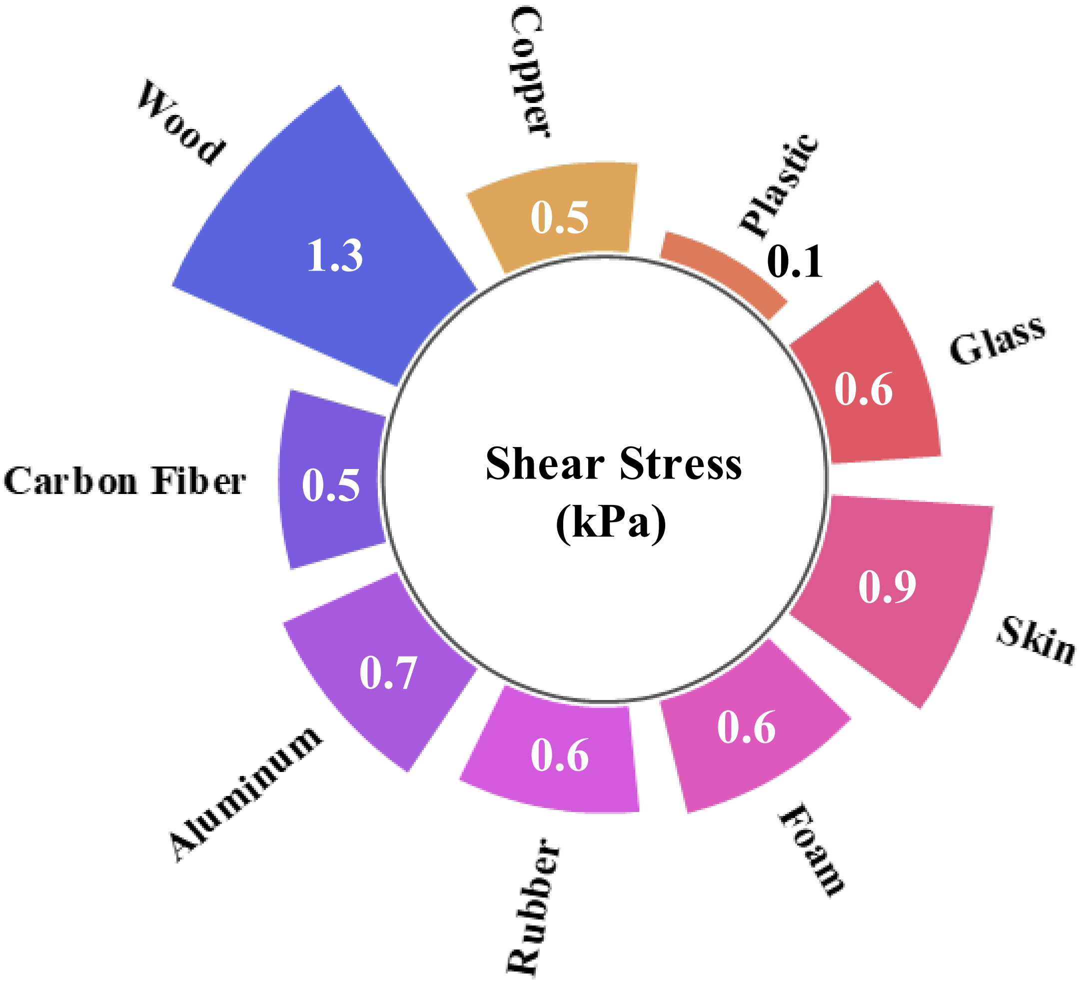

Figure S3. Surface peeling shear stress of CSP-MCG adhering to different substrates

S5. Laboratory equipment & sample sizes for sensing performance testing

In order to evaluate the effect of CSP-MCG's multifunctional characteristics, we built a series of experimental platforms and cut CSP-MCG into different shapes to cater to different usage requirements:

When testing temperature sensing characteristics of the gel, we cut CSP-MCG into a rectangular shape (as shown in FIG. S4 (a) i), and set up a temperature experiment platform (as shown in FIG. S4 (b)). Using the self-adhesive properties of the gel, attach it to the surface of the beaker (as shown in FIG. S4 (c)). The temperature of CSP-MCG is changed indirectly by heating the water beaker, and the surface temperature of CSP-MCG is measured by an infrared thermometer, and the change of resistance is recorded.

When testing the humidity sensing characteristics of the gel, we cut CSP-MCG into a square (as shown in FIG. S4 (a) ii) and changed the humidity conditions by exhalation and humidifier. The same data acquisition instrument was used to collect the resistance change signal of CSP-MCG, as shown in FIG. S4 (d)-(f).

When testing the gel strain sensing properties, we cut the CSP-MCG a long dumbbell shape (as shown in FIG. S4 (a) iii) to avoid undesirable stress concentration during the inspection.

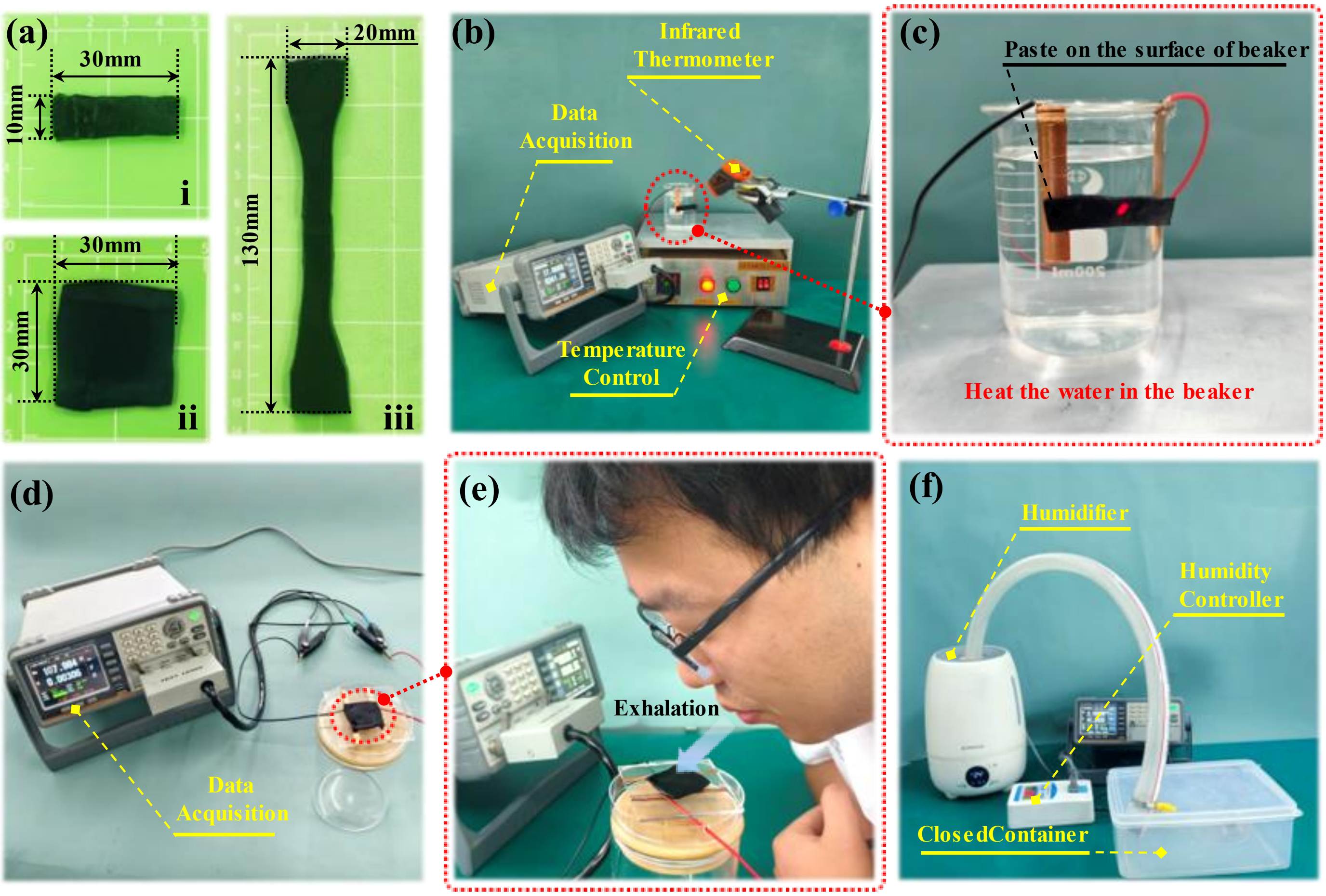

Figure S4. (a)i The CSP-MCG for temperature sensing test; (a)ii The CSP-MCG for humidity sensing test; (a)iii The CSP-MCG for tensile strain; (b)-(c) An experimental platform for testing the performance of CSP-MCG temperature sensing; (d)-(e) An experimental platform for testing the humidity sensing performance of CSP-MCG under continuous exhalation; (f) The experimental platform is built to test the humidity sensing performance of CSP-MCG under the condition of continuous step humidity change.

S6. The test results of CSP-MCG as piezoresistive material

CSP-MCG not only exhibits elasticity in tension, but also maintains good elasticity when subjected to external pressure to produce deformation. From the stress-strain curves shown in FIG. S5(a), it can be observed that the curves in the compression stage basically overlap under different magnitudes of force, and the curves in the rebound stage all return to the starting point. These experimental results indicate that CSP-MCG has stable rebound properties. Therefore, when subjected to 40% continuous compressive stress (as shown in FIG. S5(b)) , it is able to output the electrical signal stably. This reaffirms the reliable combination of elastic and conductive materials in the CSP-MCG system, which gives it the ability to change the resistance value with shape change. In addition, as a piezoresistive material, when subjected to a large external load (20 Kpa) and held for 10 s, the electrical signals remain basically unchanged as can be seen in FIG. S5(c), which exhibits good stability. It is worth noting that the response time of the CSP-MCG is less than 0.2 s at the moment of pressure, which possesses a fast response speed. FIG. S5(d) shows the functional relationship of the resistance change rate of CSP-MCG as a piezoresistive material as a function of the change in pressure magnitude, and the corresponding linear fitting curve is given. From the figure, it can be observed that the best linearity (R^2=0.99) is presented when the external pressure is high in the range of zone I. The best linearity (R^2=0.99) is observed when the external pressure is high in the range of zone II. When in the zone II, the linearity decreases slightly (R^2=0.96), probably due to the change in sensitivity. When the external pressure increases from the zone I to zone II, the sensitivity decreases from GF=5.9 to GF=1.1.

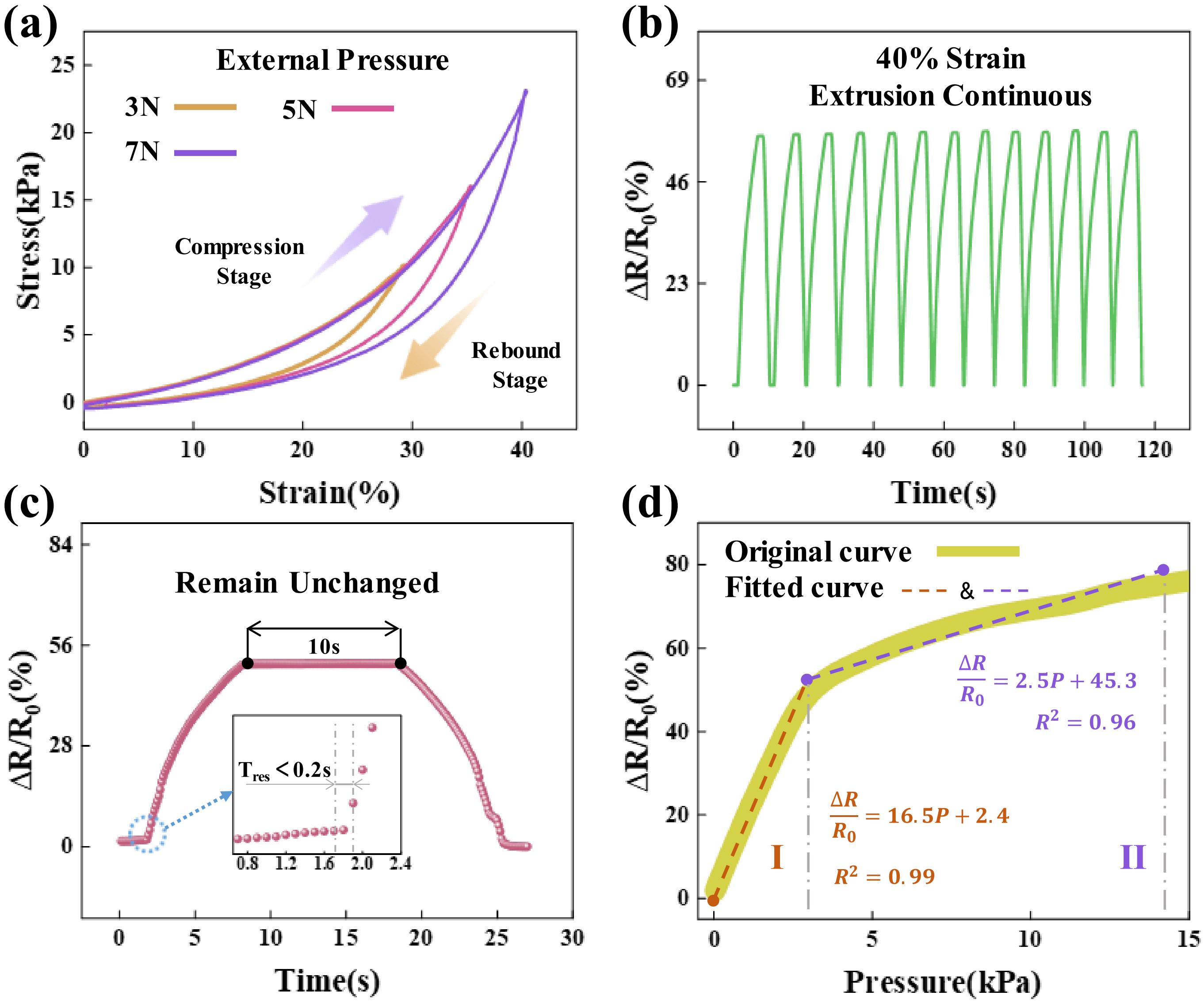

Figure S5. Piezoresistive characteristics of CSP-MCG. (a) Hysteretic curve of stress-strain under CSP-MCG compression; (b) Continuous extrusion of CSP-MCG produces an electrical signal change under 40% strain; (c) CSP-MCG as a piezoresistive material, its response time and maintaining stability diagram; (d) Linear fitting curve of resistance changes of CSP-MCG under pressure.

S7. Effect of CSP on humidity sensing of conductive gels

In order to verify whether the addition of CSP to the gel system increases the sensitivity of humidity, we prepared conductive hydrogels with only PVA as the elastic substrate (PVA-MCG) for comparison. As can be seen in FIG. S6, both PVA-MCG and CSP-MCG showed exhibit rising $\Delta R/R_0$ with increasing relative humidity. However, the sensor containing CSP shows a 5.5% increase in resistive rate of change, while the sensor without CSP shows only a 1.6% increase. Therefore, it is evident that CSP obviously improves the $\Delta R/R_0$ of the gel system under humidity sensing (more than three times). This is attributed to the natural hydrophilic property of CSP, which leads to increased gel resistance changes when adsorbing more water molecules.

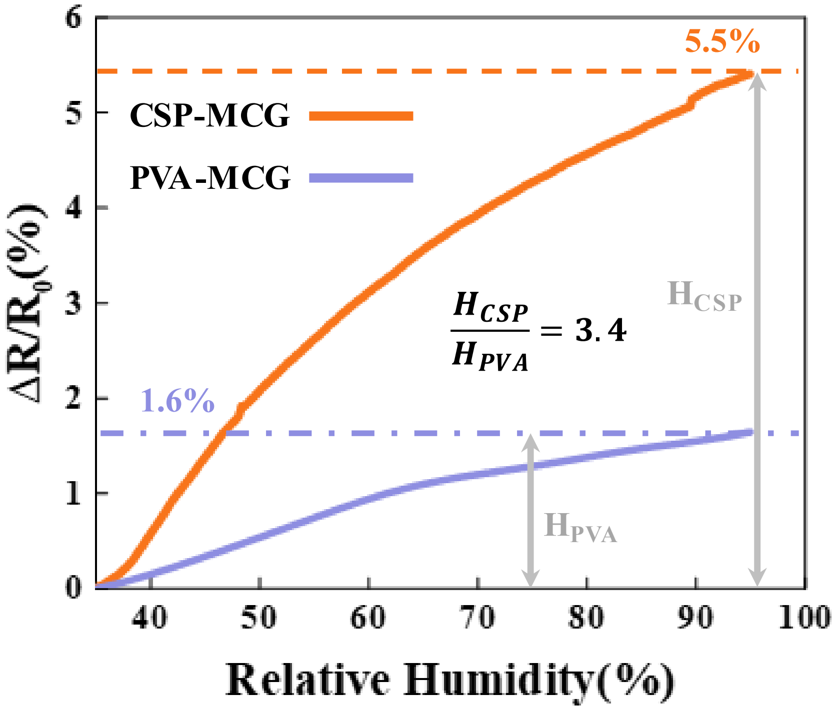

Figure S6. Comparison of resistance change between PVA-MCG and CSP-MCG with increasing relative humidity

S8. Cost comparison of common bio-based materials

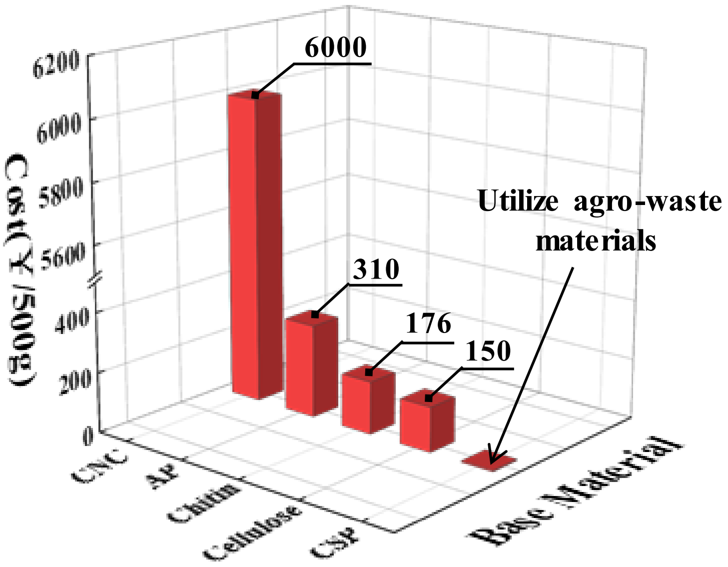

Figure S7. Comparison of the cost in RMB per 500g of common bio-based materials

S9. CSP-MCG user experience results

We attached the CSP-MCG to the skin surface of the user's face and wrist, and the experimental results showed that there was no damage to the skin during the week after wearing it. The users also feedback did not appear skin irritation.

Figure S8. Before-and-after photos of CSP-MCG users using the gel.

S10. Questionnaire of CSP-MCG users of different skin types

In order to fully evaluate the use of CSP-MCG, we conducted a sample survey of users with different skin types to provide feedback on adverse reactions and assess comfort levels. The results of sampling survey are shown in Table 3. The CSP-MCG can be directly pasted on the skin surface of users with different skin types due to the advantages of good biocompatibility of biological materials as gel substrates. No allergic or inflammatory reactions were observed. The Users have a positive evaluation of the comfort of the product.

Table 3. Summary of user experience in wearing CSP-MCG

Survey Sample	*Gender*	*Age*	*Skin Type*	*Wearing Position*	*Dermatitis or Allergy*	*Comfort Level (1 to 5)*
User 1	Male	23	Oil Skin	Face & Joints	No	4
User 2	Male	23	Dry Skin	Face & Joints	No	5
User 3	Male	24	Neutral Skin	Face & Joints	No	5
User 4	Male	22	Mixed Skin	Face & Joints	No	5
User 5	Female	25	Sensitive Skin	Face & Joints	No	4.5