Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A. This journal is © The Royal Society of Chemistry 2022

| 1 | Supporting Information |
|----|----------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 2 | |
| 3 | Ultra-Sensitive Wearable Multifunctional Flexible Sensor with Self- |
| 4 | Assembled Dual 3D Conductive Network and Yeast-Foamed Silicone |
| 5 | Rubber Foam |
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| 15 | |

16 Materials and methods

17 Materials

18 Ethylenediamine (99.5%), methyl acrylate (99%), methanol (98.5%), dimethyl sulfoxide (DMSO, 98%), N, N'-carbonyldiimidazole (99%), glucose (99%), 1-(3-19 dimethylaminopropyl)-3-ethylcarbodiimide hydrochloride (EDC, 98%), β-20 cyclodextrin (99%), N-hydroxysuc-cinimide (NHS, 98%), lithium chloride anhydrous 21 (LiCl, 99%), potassium acetate (C₂H₃KO₂, 99.9%), sodium bromide (NaBr, 99.9%), 22 potassium, (KCl, 99.8%), chloride carbon nanotube (CNT) and 1-adamantaneacetic 23 24 acid (99%) were provided by Shanghai Macklin Biochemical Co., Ltd. Dopamine hydrochloride (98%), tris(hydroxymethyl) aminomethane (Tri-Cl) (99.5%), toluene 25 (99.5%) and dimethyl sulfoxide-d6 (99.9 atom%D) was purchased from Alfa Aesar 26 27 Chemical Co., Ltd. Sulfuric acid (H₂SO₄, 96%) and nitric acid fuming (HNO₃, 95%) were provided by Shanghai Lingfeng Chemical Reagent Co., Ltd. Carbon fiber (CF) 28 and carbon ball (CB) were produced by Shanghai Lishuo Composites Co., Ltd. 29 Cellulose dialysis membrane was produced by Yuanye Biotechnology Co., Ltd. Liquid 30 silicone rubber (LSR, WSC-835) and curing agent (a mixture of ethyl orthosilicate and 31 dibutyltin dilaurate) were supplied by Jinan Xingchi Chemical Co., Ltd. Yeast powder 32 was purchased from Hubei Angel Yeast Co., Ltd. Water-resistant double-sided tape was 33 purchased from 3M China Co., Ltd. 34



35

36 Fig.S1 Synthetic route of host-guest PAMAM dendrimers.

37 Synthesis of PAMAM dendrimers and modified carbon-based fillers

G2 PAMAM and G3 PAMAM: As shown in Fig.S1, second and third-generation
polyamidoamine dendrimers were obtained by alternating reaction of ethylenediamine
and ethyl acrylate. Specific synthesis details can be found in a previous study¹.

41 **CF-CD:** The carbon fiber (CF) was ultrasonically cleaned with acetone at 60 °C for 42 6 h, washed with deionized water until the acetone was completely removed, and then 43 dried at 100 °C. The acid oxidation process uses a 3:1 (v/v) mixture of concentrated 44 H_2SO_4/HNO_3 and sonicates for 45 min at room temperature. Then it was washed with 45 deionized water to pH=7. It was dried at 100 °C to obtain acid oxidized carbon fiber 46 (CFs). The DMSO solution (10 mL) containing β -cyclodextrin (6.04 g, 5.37 mmol) was

mixed with the DMSO solution (15 mL) of N, N'-carbonyldiimidazole (8.62 g, 53.67 47 mmol). After stirring for 6 h, a DMSO solution (10 mL) of G3 PAMAM (third-48 generation polyamide-amine dendrimer, 2.00 g, 0.20 mmol) was added dropwise to the 49 above mixture solution. After 72 h, the reaction solution was dialyzed with water (12 50 times, 3 L) for 3 d using a dialysis membrane with an MWCO of 3500. A white powder 51 of the G3 PAMAM-CD (host dendrimer) product was obtained by a further freeze-52 drying process. The carbon fiber (CFs, 0.50 g) was completely immersed in the DMSO 53 solution, and the internal air bubbles were completely removed with the aid of a 54 vacuum. Then, EDC (0.62 g, 3.23 mmol) and NHS (0.77 g, 3.23 mmol) were added to 55 the above DMSO solution and sonicated for 30 min at room temperature. Then, 20 mL 56 DMSO solution containing G3 PAMAM-CD (0.50 g) was added. The amount of 57 grafted G3 PAMAM-CD was controlled by ultrasonic for 30 min, 6 h and 24 h at room 58 temperature. Finally, the carbon fiber was dialyzed with water (20 times, 10 L) for 3 d 59 with a dialysis membrane and a cut-off (MWCO) of 7000, and the water was removed 60 by a freeze dryer. Thus, the CF-CD was obtained. 61

CB-Ad: The carbon balls (CB) were ultrasonically cleaned with acetone at 60 °C for 63 6 h, washed with deionized water until the acetone was completely removed, and then 64 dried at 100 °C. The acid oxidation process uses a 3:1 (v/v) mixture of concentrated 65 H₂SO₄/HNO₃, and ultrasonics at 70 °C for 12 h. Then it was washed with deionized 66 water to pH=7. It was dried at 100 °C to obtain acid oxidized carbon balls (CB-COOH). 71 The G2 PAMAM-Ad (guest dendrimer) dendrimer was synthesized by EDC/NHS 72 coupling chemistry. EDC (2.47 g, 12.91 mmol) and NHS (1.48 g, 12.91 mmol) were

added to the DMSO (15 mL) solution containing 1-adamantaneacetic (0.25 g, 1.32 69 mmol) and stirred at 25°C. After 30 min, the mixture was added dropwise to a DMSO 70 (10 mL) solution containing G2 PAMAM (second-generation polyamide-amine 71 dendrimer, 1.00 g, 0.81 mmol), and stirred vigorously at room temperature for 3 d. 72 Using a dialysis membrane with a molecular weight cut-off (MWCO) of 1000, the 73 reaction mixture was dialyzed with water (20 times, 10 L) for 3 d. G2 PAMAM-Ad 74 was obtained as a white powder after a further freeze-drying process. The CB-COOH 75 was dispersed in DMSO (2 mg/mL) and dispersed through an ultrasonic bath. Then, 76 77 300 mL of CB-COOH solution was mixed with 330 mL of EDC/NHS (5 mg/mL, EDC/NHS 1:1) solution, and the mixture was sonicated at room temperature for 60 min. 78 The mixture was added dropwise to 20 mL G2 PAMAM-Ad solution (0.1 g/mL) and 79 80 sonicated at room temperature for 24 h to separate CB-COOH for the reaction. The dispersion was dialyzed with water (20 times, 10 L) for 3 d using a dialysis membrane 81 with a cut-off (MWCO) of 3500, and the water was removed by a freeze dryer. The 82 83 CB-Ad was thus obtained.



Fig.S2 Schematic diagram of the synthetic route and interaction of (a) internal network CF-CD/CBAd and (b) external network PDA-GO/CNT-COOH.

PDA-GO: 100 mL of 10 Mm Tris-Cl solution (pH=8.5) was prepared. GO (100 mg) and dopamine hydrochloride (50 mg) were added to the above solution and dispersed by sonication for 30 min in an ice bath. After stirring for 24 h at 70 °C, the mixture was centrifuged (10000 rpm) and washed 6 times with distilled water. Finally, PDA-GO was obtained by filtration and dried under reduced pressure at 60 °C for 24 h.

92 **CNT-COOH:** The carbon nanotubes (CNT) were ultrasonically cleaned with 93 acetone at 60 °C for 6 h, washed with deionized water until the acetone was completely 94 removed, and then dried at 100 °C. The acid oxidation process uses a 3:1 (v/v) mixture 95 of concentrated H_2SO_4/HNO_3 , and ultrasonics at 70 °C for 12 h. Then it was washed 96 with deionized water to pH=7. It was dried at 100 °C to obtain acid oxidized carbon 97 nanotubes (CNT-COOH).

98 Preparation of SF with different pore size

Yeast (0.1 g), glucose aqueous solution (1 mL, 1 g/mL) and curing agent (0.4 g) were 99 added to the LSR, and then placing the mixture in incubator at 28, 34 and 40 °C to cure 100 for 24 h. Yeast respiration and the slowly curing process of LSR made the cured silicone 101 rubber foam (SF) composites have a porous structure, and then placed SF in a 50 °C 102 aqueous solution ultrasonic for 1 h to remove glucose and yeast. After drying in an oven 103 at 40 °C for 6 h, the SF-M (28 °C, average pore size is 457 µm), SF-L (34 °C, average 104 pore size is 621 µm) and SF (40 °C, average pore size is 194 µm) were obtained (Int-105 SF). 106

107 Characterizations

Through fourier transform infrared spectroscopy (AVATAR 370, Nicolet, USA), 108 Raman spectroscopy (Horiba Evolution, 633 nm, France), X-ray photoelectron 109 spectroscopy (Thermo Kalpha, USA) and hydrogen nuclear magnetic resonance 110 spectroscopy (Bruker Avance-III 400 MHz, Switzerland) studied the chemical 111 structures of dendrimers, PDA-GO, CNT-COOH, CF-CD, CB-Ad and composites. 112 OM, SEM and TEM images were obtained using an optical microscope (OM, XPF-113 300, Japan), scanning electron microscopy (Hitachi S4800, Japan) and transmission 114 electron microscope (FEI Tecnai G2 F20, USA), respectively. A universal testing 115 machine (Instron 5966, USA) was used to test mechanical properties. Keithley DMM 116 117 2450 was used to detect electrical signals of sensors. The temperature sensing process of Dual-SF was recorded using an infrared camera (FLIR, DALI T8, China), The 118 human wearable tests were carried out after the experimenter signs a written informed 119 consent. The I-V curves were measured using an electrochemical workstation 120

(CHI660E, USA). The humidity sensing response of the sensor was measured using a
saturated salt solution as the humidity source. By dissolving LiCl (RH 11%), C₂H₃KO₂
(RH 23%), NaBr (RH 57%) and KCl (RH 84%) in deionized water in an airtight glass
container at 25 °C.

Stretch sensing measurements: The tensile sensing characteristics of the foam 125 composites were measured using a digital multimeter (Keithley DMM 2450). First, 126 fixed the two ends of the different foam composites (Int-SF, Ext-SF and Dual-SF) with 127 a stripe size of 60 mm \times 15 mm \times 3.2 mm on the tensile testing machine (Instron 5966) 128 fixture, and then stretched. The resistance change of the different foam composites were 129 read by the computer connected to the multimeter. The stability of Dual-SF was also 130 measured by 1000 cycles of stretch-release (elongation is 40%). The average stretching 131 speed was 5 mm/min. 132

Wearable testing measurements: The wearable test of strip Dual-SF and meshed 133 Dual-SF were also measured using Keithley DMM 2450. The strip Dual-SF were fixed 134 to the body parts such as fingers (60 mm \times 15 mm \times 3.2 mm), wrist (60 mm \times 15 mm 135 \times 3.2 mm), elbows (60 mm \times 15 mm \times 3.2 mm) and knee (180 mm \times 30 mm \times 3.1 mm) 136 with insulating tape. The meshed Dual-SF were fixed to the arm muscles, leg muscles, 137 138 neck and wrist with insulating double-sided tape. The resistance change caused by the movement of the human body was read by the computer connected to the multimeter. 139 To avoid errors caused by temperature changes, all wearable tests started after the 140 sensor temperature was the same as the body temperature (i.e., the resistance of the 141 sensor was stable). 142

Pressure sensing measurements: The change in resistance of the (Int-SF, Ext-SF 143 and Dual-SF in response to pressure was measured by a digital multimeter (Keithley 144 DMM 2450) and data acquisition was performed by a computer connected to the digital 145 multimeter. The resistance changes of Dual-SF in the process of pressing the keys of 146 the keyboard, rigid ceramic and elastic rubber ball were tested by using a strip sample 147 with a size of 20 mm \times 20 mm \times 3.2 mm. Changes in resistance during different 148 amplitudes of motion were tested through a Dual-SF (100 mm \times 80 mm \times 3 mm) placed 149 under the sole. The size of a single sensor assembly in the pressure sensing array is 15 150 $mm \times 15 mm \times 3 mm$. All tests involving body part compressions were performed in 151 an incubator (36 °C) to avoid the interference of temperature. 152

Temperature sensing measurements: The resistance changes of the sensors in 153 154 response to temperature were measured by a digital multimeter (Keithley DMM 2450), and data acquisition was performed by a computer connected to the digital multimeter. 155 Using a hot plate to control the measured temperature of the Dual-SF/LSR sensor (30 156 mm \times 25 mm \times 0.9 mm). Real-time resistance at 25 °C \sim 95 °C was recorded. The body 157 temperatures of the hands and arms were continuously monitored for 90 S in the 158 morning and evening. The temperatures of the neck were also monitored for 60 S before 159 160 and after sport. The temperatures of mainframe temperature were monitored for 5 S during the stable operation of the computer. All changes in Dual-SF resistance caused 161 by temperature fluctuations were recorded. An infrared (IR) camera (FLIR, DALI T8, 162 China) was used for the real-time recording of infrared images of the test process. 163



166 Fig.S3 ¹H-NMR of (a) G2 PAMAM-Ad (guest dendrimer) and (b) G3 PAMAM-CD (host

167 dendrimer); (c) FTIR spectra of G2 PAMAM-Ad and G3 PAMAM-CD.





169

170 Fig.S4 TEM images of (a) GO and (b) PDA-GO.



172

173 Fig.S5 (a) Raman spectra of different carbon-based fillers. (b) XPS spectra of different carbon-based

- 174 fillers. Deconvoluted C1s XPS spectra of (c1) PDA-GO, (c2) CF-CD and (c3) CB-Ad. (d) FTIR
- 175 spectra of SF and Dual-SF.



177

- 178 Fig.S6 Optical microscopy images of (a) SF and (b) CF-CD/CB-Ad interconnected network
- 179 structures.



180

181 Fig.S7 Cyclic rubbing test of (a) Dual-SF and (b) Int-SF/GO/CNT-COOH.



Fig.S8 Schematic of cracked pore walls improving the continuity of the conductive network.



187 Fig.S9 SEM images of (a) Int-SF/PDA-GO/original CNT and (b) Dual-SF.



190 Fig.S10 Steel ball rebound test indicating the elasticity of the Dual-SF.

As shown in Fig.S10, the high elasticity of the Dual-SF foam structure was further verified by the steel ball rebound experiment, and the resilience rate (RT) was introduced to characterize the elasticity of the Dual-SF foam. The quantification formula is as follows:

$$195 \quad RTa = Ha/H0 \tag{1}$$

Where *H0* is the initial height, and Ha is the height of the steel ball after rebounding
once or twice. The steel ball fell freely onto the surface of Dual-SF from a height of 15
cm, and rebound quickly after a collision. In two consecutive rebound experiments, the
RT of Dual-SF were 86% and 57%, respectively.

200



201

202 Fig.S11 (a-c) SEM images of SF with different pore sizes. (d) Pore size and distribution of SF,

203 SF-M and SF-L. (e) Mechanical properties of SF, SF-M and SF-L (compressive strain is 70%).

205 Table.S1 Comparison of pore diameter and density of silicone rubber foams prepared at different

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206 temperatures.
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| Materials | Temperature (°C) | Pore diameter (µm) | Density (g/cm ³) |
|-----------|------------------|--------------------|------------------------------|
| SF-M | 28 | 457 | 0.227 |
| SF-L | 34 | 621 | 0.143 |
| SF | 40 | 194 | 0.356 |



209 Fig.S12 Changes in the internal conductive network of Dual-SF during stretching process.



212 Fig.S13 Repeatability of resistance change at (a) 0.2% and 0.5% and (b) 10%, 20% and 50% strain.

213 (c) The change of Dual-SF resistance during the process of high leg lifts and push-ups exercise.

214



216 Fig.S14 (a) Finite element analysis of stress distribution of meshed Dual-SF under external force.

217 Changes in the resistance of (b) meshed Dual-SF and (c) strip Dual-SF during forearm muscle

218 contraction.

219

| Materials | Gauge factor | Strain (%) | Ref. |
|-------------------------|--------------|------------|-----------|
| | 3.42 | 0-50 | |
| MXene hydrogels | 4.77 | 50-100 | 2 |
| | 5.82 | 100-180 | |
| NACA as AAm/LiCl | 0.44 | 0-150 | 2 |
| NAGA-co-AAm/LiCl | 0.94 | 150-300 | 3 |
| VCDD/CNIT | 4.24 | 0-170 | 4 |
| ASBK/CN1 | 25.98 | 170-214 | 4 |
| NR/graphene | 46 | 0-3 | 5 |
| | 1.66 | 0-200 | |
| (EMIM)Cl ionohydrogels | 3.64 | 200-600 | 6 |
| | 6.78 | 600-800 | |
| | 1.79 | 0-100 | |
| MXene/HK hydrogel | 5.27 | 100-500 | 7 |
| | 10.22 | 500-1000 | |
| PU-DA-1/1-PANI hydrogel | 2.89 | 0-120 | 8 |
| | 1.09 | 0-30 | |
| POSS-TMB-LiMTFSI | 2.45 | 30-60 | 9 |
| | 7.03 | 60-100 | |
| | 15 | 0-44 | |
| TPU/CNT | 30 | 44-73 | 10 |
| | 58 | 73-100 | |
| | 26.07 | 0-0.6 | |
| | 5.71 | 0.6-10 | |
| (CNF)/AgNWs/CA@PU | 1.58 | 10-30 | 11 |
| | 0.52 | 30-70 | |
| | 16.95 | 0-28 | |
| Int-SF | 96.82 | 28-60 | |
| | 43.72 | 0-32 | |
| Ext-SF | 137.54 | 32-41 | This work |
| D 1.77 | 128.91 | 0-45 | |
| Dual-SF | 690.76 | 45-70 | |

Table.S2 Comparison of the gauge factor of Dual-SF and the reported flexible sensors²⁻¹¹.



223 Fig.S15 Electrical model of Dual-SF at different pressures.





226 Fig.S16 *I-V* curves of the (a) Int-SF, (b) Ext-SF and (c) Dual-SF under different applied pressures.



229 Fig.S17 Comparison of pressure sensitivity of Dual-SF and the reported pressure sensors.

228



232 Fig.S18 (a) The change of Dual-SF resistance during the process wrist pulse. (b) Transient

233 responses of loading and removal of different numbers of petals. (c) Response/recovery time of

234 Dual-SF under 10 kPa, 20 kPa, 30 kPa and 50 kPa.

| | Pressure sensitivity | Pressure ranges | Df |
|------------------------------|----------------------|-----------------|-----------|
| Materials | (kPa ⁻¹) | (kPa) | Ket. |
| | 20.3 | 0 - 1 | |
| PDA-CNT/SR foam | 15.1 | 1 - 2.5 | 12 |
| | 0.6 | 5 - 12.5 | |
| | 59.0 | 0 - 2 | 12 |
| GNWS/PDMS | 4.8 | 2 - 20 | 13 |
| | 54.31 | 0 - 0.5 | |
| T ' 1 | 30.11 | 0.5 - 10 | 14 |
| Ionic gel | 8.42 | 10 - 40 | 14 |
| | 1.03 | 40 - 115 | |
| | 19.32 | 0 - 0.25 | 15 |
| PPy Film/PDMS | 0.51 | 0.25 - 2 | 15 |
| | 134 | 0 - 1.5 | |
| AgNWs/GR/PANFs | 3.5 | 1.5 - 15 | 16 |
| | 0.7 | 15 - 75 | |
| | 4.05 | 0 - 1 | 17 |
| MXene/rGO aerogel | 22.56 | 1 - 3.5 | 17 |
| PLGA/PVA/NF film | 6.33 | 0.3 - 11.6 | 18 |
| CNTs/Ni-fabrics | 26.13 | 0.2 - 982 | 19 |
| | 0.55 | 0.23 - 0.982 | |
| MXene/PLA sheet | 3.81 | 0.982 - 10 | 20 |
| | 2.52 | 10 - 30 | |
| | 57.5 | 0 - 0.75 | 21 |
| AgNPs hydrogel/Cu electrodes | 171.4 | 0.75 - 1.5 | 21 |
| | 169.94 | 0 - 13.1 | |
| Dual-SF | 108.41 | 13.1 - 34.2 | This work |
| | 7.02 | 34.2 - 104.0 | |

| 236 | Table.S3 Comparison of p | ressure sensitivity of Dual-SF | and the reported pressure s | sensors(ref ¹²⁻²¹). |
|-----|--------------------------|--------------------------------|-----------------------------|---------------------------------|
|-----|--------------------------|--------------------------------|-----------------------------|---------------------------------|





238 Fig.S19 Comparison of response/recovery time of Dual-SF (thickness is about 0.9 mm) and Dual-

239 SF/LSR (thickness is about 0.9 mm) to temperature change (30-85 °C).

241 **Table.S4** Comparison of the temperature sensing performance of our Dual-SF and the reported

242 sensors²²⁻³².

| Materials | TCR (%/°C) | Sensing ranges (°C) | Ref. |
|------------------------------------|------------|---------------------|-----------|
| PNIPAAm/PVA/SA | 0.58 | 20 - 40 | 22 |
| PVA/Gly/CNT/CB | 0.935 | 30 - 90 | 23 |
| MXene-Ti3C2Tx@EDOT | 0.86 | 26 - 40 | 24 |
| rGO | 0.801 | 25 - 95 | 25 |
| PHB/rGO | 0.8 | 20 - 65 | 26 |
| Polymine-AgNPs | 0.17 | 24 - 54 | 27 |
| PEDOT:PSS | 0.03 | 30 - 45 | 28 |
| Laser-engraved graphene | 0.06 | 25 - 50 | 19 |
| Graphene-MPPU | 0.815 | 20 - 100 | 30 |
| Pt NFs into silk fibroin membranes | 0.205 | 20 - 60 | 31 |
| BP/LEG on SEBS | 0.1736 | 25 - 50 | 32 |
| Int-SF/LSR | 0.308 | 30 - 100 | |
| Ext-SF/LSR | 1.143 | 30 - 100 | This work |
| Dual-SF/LSR | 1.621 | 30 - 100 | |

Table.S5 The skin and mainframe temperatures measured by infrared camera were compared with the temperatures calculated by the resistance-temperature curve. (The skin temperature measured by infrared thermal imaging is the average of 10 measurements, with a 10 s interval between each measurement.)

| Position | Situation | Temperature (°C, Infrared thermal imager) | Temperature (°C, Thermal induction curve calculation) | Error (°C) |
|--------------------|---------------|-------------------------------------------------|-------------------------------------------------------------|---------------|
| Hand | Morning 1 | 34.2 | 34.4 | +0.2 |
| Hand | Evening 1 | 35.9 | 36.3 | +0.4 |
| Arm | Morning 2 | 34.1 | 34.5 | +0.4 |
| Arm | Evening 2 | 36.8 | 36.7 | -0.1 |
| Neck | Original | 35.1 | 35.1 | 0 |
| Neck | After sport 1 | 37.2 | 37.3 | +0.1 |
| Neck | After rest | 35.8 | 35.7 | -0.1 |
| Neck | After sport 2 | 37.5 | 37.6 | +0.1 |
| Mainframe shell | 1.5 s | 56.9 | 56.3 | -0.6 |
| Mainframe shell | 3 s | 70.7 | 71.1 | +0.4 |



Fig.S20 (a) Dynamic responses of the Dual-SF to the relative humidity (RH) from 11 to 84%. (b) Analyses of the response and recovery time in the detection of 84% RH. (c) Signal comparison when pressing Dual-SF with wear gloves and bare fingers. (d) Change in resistance of Dual-SF directly attached to the skin during flexion of the inside of the wrist. (e) Change in resistance of Dual-SF attached to rubber gloves during flexion of the inside of the wrist. (f) Change in resistance of Dual-SF attached to the skin by double-sided tape during flexion of the inside of the wrist (the initial resistance when not in contact with the skin was defined as R_0).

The humidity sensing response of the sensor was measured using a saturated salt solution as the humidity source. By dissolving LiCl (RH 11%), $C_2H_3KO_2$ (RH 23%), NaBr (RH 57%) and KCl (RH 84%) in deionized water in an airtight glass container at 260 25 °C. Fig.S20a shows that the resistance of the Dual-SF sensor slightly increased with increasing humidity, which was attributed to the adsorption of H₂O molecules by PDA-

GO/CNT-COOH. The porous structure of the sensors increased the contact area of the 262 surface-coated PDA-GO/CNT-COOH with water vapor, and the PDA-GO/CNT-263 COOH had a high density of surface vacancies and polar groups. Therefore, PDA-264 GO/CNT-COOH can easily capture H_2O molecules in the environment³³. The charge 265 transport of PDA-GO and CNT-COOH exhibits p-type semiconductor behavior 266 dominated by positive charge carriers (holes). The H₂O molecules captured by PDA-267 GO/CNT-COOH acted as electron donors, reducing the charge carrier concentration 268 and increasing the resistance of the sensor^{34, 35}. The response/recovery was defined as 269 the signal to change 90% of the peak value. Fig.S20b shows that the response/recovery 270 time of Dual-SF was 6.7 s and 25.1 s, respectively. 271

We also study the effect of humidity on the strain sensing performance of the sensor. 272 Fig.S20c shows the compression signal of bare fingers was slightly lower than that of 273 wearing gloves, which was attributed to the increased resistance caused by the humidity 274 of the finger. In order to avoid the interference of humidity on the sensing signal, we 275 adopted different solutions. In the wearable tests (Fig.2f-h, S13c and S18a), Dual-SF 276 was not in direct contact with the skin but was fixed on the body by a water-resistant 277 double-sided tape. As shown in Fig.S20d-f, the average value of the sensor resistance 278 in direct contact with the skin was significantly higher than that in the case of wearing 279 gloves. However, the average value of the sensor resistance change separated by the 280 double-sided tape was the same as wearing rubber gloves. This proved that the double-281 sided tape can effectively avoid the rise in sensor resistance caused by body humidity. 282 In addition, rubber gloves can also effectively block moisture from the skin. In the 283

pressing tests (Fig.3d-f and i), we mainly adopted this method to avoid the signal interference caused by humidity. In particular, the meshed Dual-SF was directly attached to the skin surface in the wearable test. This is because the large areas covered with double-sided tape will increase the discomfort of the actual wearable. The hollow design improved signal strength and provided better breathability. We think this reduced some skin moisture disturbances. Furthermore, all relevant tests were carried out after the signal was stable, which further avoided test errors caused by humidity.





Fig.S21 (a1) Finite element analysis of stress distribution at cracks under tension. (a2) Comparison
of resistance changes during stretching of original Dual-SF and ruptured Dual-SF (elongation is
20%). (b1) Finite element analysis of stress distribution at cracks under compression. (b2)
Comparison of resistance changes during compression of intact Dual-SF and ruptured Dual-SF (10

kPa). Changes in pressure sensing performance before and after simulating sweat immersion in the
(c1) Int-SF, (c2) Ext-SF, (c3) Dual-SF (20 kPa).

The effects of rupture and sweat intrusion on the sensing performance of Dual-SF 299 were also investigated. Fig.S21a1 shows that stress concentration occurred at the Dual-300 SF crack during the stretching process and induced larger deformation, which led to 301 more severe damage to the conductive network inside the sensor. Fig.S21a2 also shows 302 a larger change in resistance of Dual-SF with cracks at the same elongation ratio. 303 However, the cracks of Dual-SF gradually extended under tension. The resistance 304 changes of the cracked Dual-SF gradually increased with the increase of cycling, and 305 the resistance cannot be recovered after unloading the stress. Fig.S21b1 shows that the 306 cracks closed and generated stress concentration under pressure, which made more 307 conductive paths inside the sensor. Fig.S21b2 shows that the resistance change of Dual-308 SF with cracks was more significant. This was also consistent with the above sensing 309 mechanism. However, the uncertainty of the mutual contact of the fractured surfaces 310 311 resulted in the unstable resistance change of the Dual-SF.

The simulated sweat solution (0.03 mL, aqueous solution containing 0.5 wt.% NaCl, 0.1 wt.% lactic acid and 0.1 wt.% urea) was dropwise to the glass plate, a 15 mm × 15 mm × 3 mm sensor was covered on the droplet. Observed the output signal of the sensor during the cyclic pressing process. In the original state, Int-SF, Ext-SF and Dual-SF can output clear and stable signals for pressure changes (Fig.S21c). After immersion in the simulated sweat solution, the Int-SF signal basically disappeared (Fig.S21c1). This was because the pure silicone rubber pore walls of Int-SF were impermeable to water, and the droplets bound together inside the Int-SF, resulting in short circuits and signal
crosstalk³⁶. In contrast, the large number of polar groups carried by PDA-GO and CNTCOOH endowed the pore walls of Ext-SF and Dual-SF with good hydrophilicity.
During the pressing process, the droplets can spread around in the Ext-SF and Dual-SF,
thus preventing the droplets from agglomerating³⁷. Fig.S21c2 and c3 show that droplet
immersion had less effect on the sensing performance of Ext-SF and Dual-SF.

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