Supporting Information

Sensing Arbitrary Contact Forces with a Flexible Porous Dielectric Elastomer

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Detailed Description of the Device Fabrication and Methods

The Electrode Fabrication

The PDiF device was fabricated by sandwiching a porous elastomer between two textile electrode layers. The patterned electrodes were prepared by a home-made printing procedure. As illustrated in Figure S1a, a piece of cotton with the thickness of 150 μm was covered by a tape layer (50 μm in thickness, 3M). Direct laser machining (VLS 2.30, Universal Laser) was used to cut through the tape layer with a pre-designed geometrical pattern. After the tape in the unwanted areas was peeled off, the rest served as a stencil mask for printing (Figure S1b). Silver paste ink with a volume resistivity of 100 mΩ•mm was applied onto the cotton/tape substrate surface (Figure S1c). Subsequently, a squeegee was used to spread the ink over the surface, and force it into the cotton substrate through the openings of the tape (Figure S1d). The substrate was warmed by a heat gun (SAIKE 852D, power 100 W) for 20 s. After the silver ink was completely dried, the rest tape was removed from the cotton surface, leaving the electrode of cotton/silver ink composite (Figure S1e).

Preparation of the Porous Dielectric

Sodium dodecyl sulfate (SDS, Sigma-Aldrich) and lithium sulfate (Li_2SO_4, Sigma-Aldrich) were mixed into deionized (DI) water (SDS: Li_2SO_4: DI = 1:20:100, w/w). The SDS/Li_2SO_4/DI (SLD) mixture (Figure S2a) was treated by shaking for 20 min in a vortex shaker (VORTEX-5, Kylin-Bell). Next, the as-prepared SLD mixture was separately added to the two components (part A and B) of Ecoflex-0030 (SMOOTH-ON) with a mass ratio of 1:1 under a manual stirring for 5 min (Figure S2b). They were mixed a beaker (1:1, w/w) and thoroughly stirred for another 5 min. The final mixture containing SLD, part A and part B of Ecoflex-300 (abbreviated as the Ecoflex mixture) was de-gassed for 15 min in a vacuum chamber (Figure S2c).

Sensor Assembly

As shown in Figure S2d, two pieces of U-shape acrylic molds were placed on the bottom electrode surface, forming a rectangular ring-shape chamber. The Ecoflex mixture was poured and filled in the chamber, followed by covering the upper electrode layer (Figure S2e, S2f). The sample was cured at room temperature for half an hour, before the removal of the acrylic molds (Figure S2g). Finally, the whole device was placed in a room condition for three days. During this procedure, the component of water was completely evaporated, leaving plenty of pores in the elastomer.

Force Measurement and Mathematic Processing

The performance of the PDiF sensors were evaluated with a customized setup consisting of a dynamometer (M5-05, Mark-10) with a resolution of 1 mN and a linear stage platform (LMS, LTS300/M, Thorlabs Inc.) with a resolution of 1 μm. The setup can control and monitor mechanical loads and displacements simultaneously. To apply a uniform normal force on each unit of the PDiF sensor, an acrylic sheet with a thickness of 1 mm was glued with the sensor. The normal force was directly applied on the sheet surface. For the calibrations on the tangential responses, a force in the direction parallel to the sensor surface was applied to the acrylic sheet. The experimental setup was illustrated in Figure S3 for the calibrations on a force with an arbitrary direction. A weight was placed on the sheet surface, serving as the normal load. The acrylic sheet was cut into polygon shape with specific angles, in which the force gauge can generate a parallel force with a designed in-plane angle. The corresponding capacitance change in each sensing unit was measured by a LCR meter (WK 65120B) or a customized circuit at an AC excitation voltage of 1 V and a frequency of 10 kHz.

The consents of all volunteers have been obtained for participations in all the experiments.

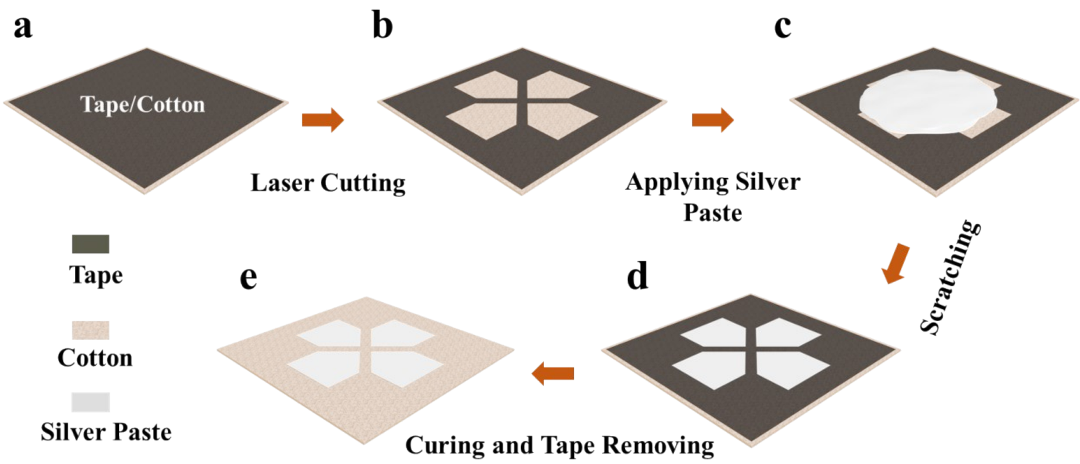

Figure S1 Electrode Fabrication. (a) Laminate a layer of one-side tape on the cotton substrate. (b) Laser cut on the tape layer to define the geometrical patterns. (c) Apply silver paste on the surface of the substrate. (d) Squeegee the silver paste into the patterns of the tape layer. (e) Solidify the silver paste and remove the tape on the substrate.

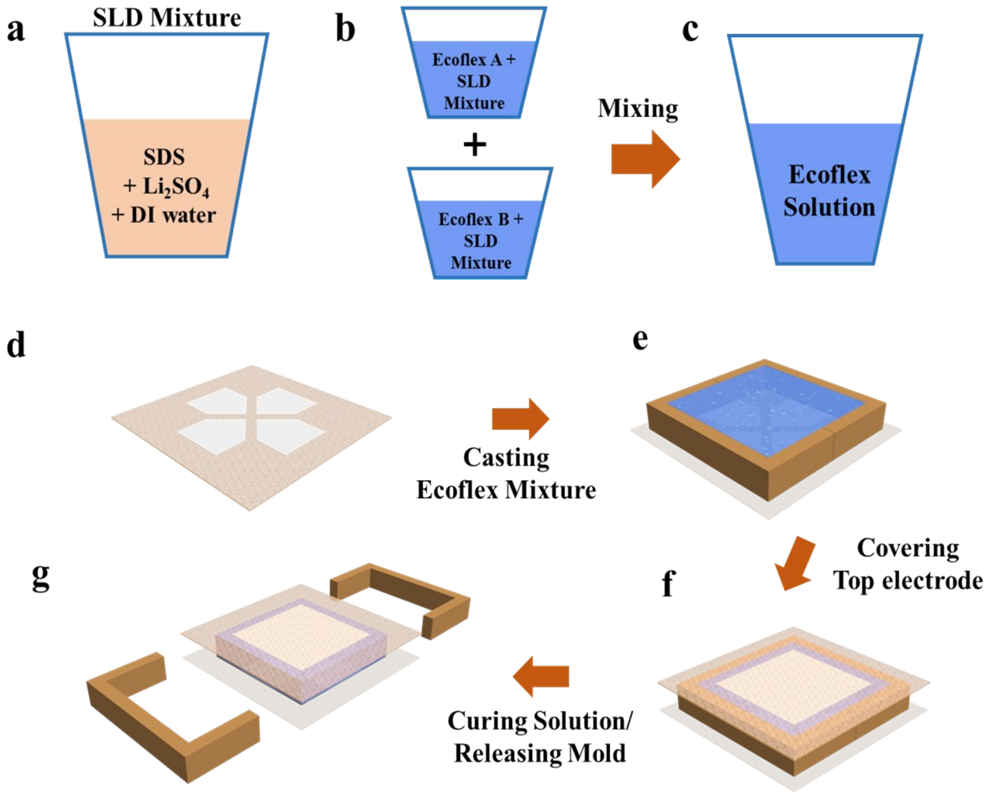

Figure S2 Device Fabrication. (a) Prepare the SLD solution by mixing SDS, Li_2SO_4 and DI water at a mass ratio of 1:20:100. (b-c) Prepare the Ecoflex mixture by adding the SLD solution into the two components (part A and part B) of Ecoflex separately, and mixing them together. (d-e) Cast the Ecoflex mixture into a molded reservoir on the electrode surface. (f) Cover the top electrode layer on the Ecoflex solution. (g) Solidify the Ecoflex mixture and release the mold of the reservoir.

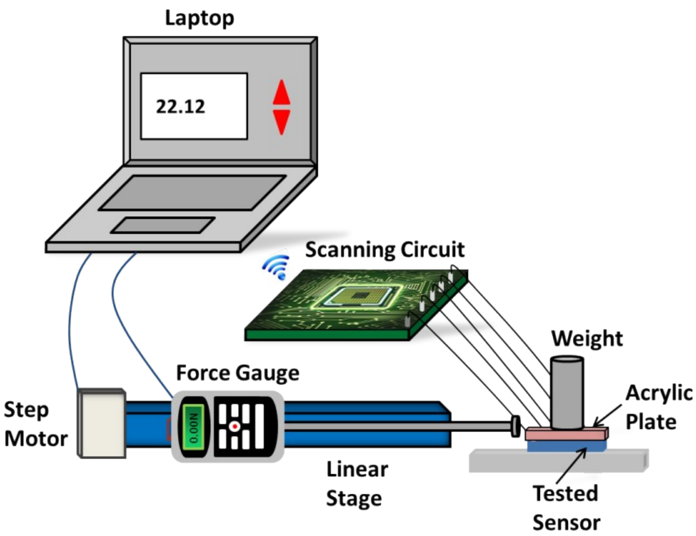

Figure S3 Schematic illustration on the experimental setup of the device characterization on the force in an arbitrary direction**.**

The Relationship of Capacitive Outputs and Three-Dimentional Forces

We have developed a coefficient matrix A to investigate the transduction principle between the spatial forces and the capacitance outputs. The relative capacitance changes, $\Delta C_1/C_1^0$, $\Delta C_2/C_2^0$, $\Delta C_3/C_3^0$, $\Delta C_4/C_4^0$, and the force components, F_x, F_y, F_z, follow Equation (2).

The cofficient matrix A is expressed as:

$$A = \begin{bmatrix} \alpha_{11} & \alpha_{12} & \alpha_{13} & \beta_{11} & \beta_{12} & \gamma_{11} & \gamma_{12} \\ \alpha_{21} & \alpha_{22} & \alpha_{23} & \beta_{21} & \beta_{22} & \gamma_{21} & \gamma_{22} \\ \alpha_{31} & \alpha_{32} & \alpha_{33} & \beta_{31} & \beta_{32} & \gamma_{31} & \gamma_{32} \\ \alpha_{41} & \alpha_{42} & \alpha_{43} & \beta_{41} & \beta_{42} & \gamma_{41} & \gamma_{42} \end{bmatrix} \quad \text{(S1)}$$

where the coefficients, α_{ij} (i = 1, 2, 3 or 4 and j = 1, 2 or 3) represent the linear relationship between relative capacitance changes and the three force components. In addition, due to the presence of the porous structure, the inputs and outputs show a nonlinear relationship, governed by β_{ij} and γ_{ij}.

According to the experimental measurements of ($\Delta C_1/C_1^0$, $\Delta C_2/C_2^0$, $\Delta C_3/C_3^0$, $\Delta C_4/C_4^0$, F_x, F_y, F_z) with a total number of n = 167, we have determined the coefficients of α_{ij}, β_{ij} and γ_{ij} by using least squares algorithm. Equation (3) gives the coefficient matrix and Figure S4 compares the experimental data (blue dots) and fitted values (red dots) calculated from Equations (4-6). The root mean squared error is 0.033 between the experiment and fitted data.

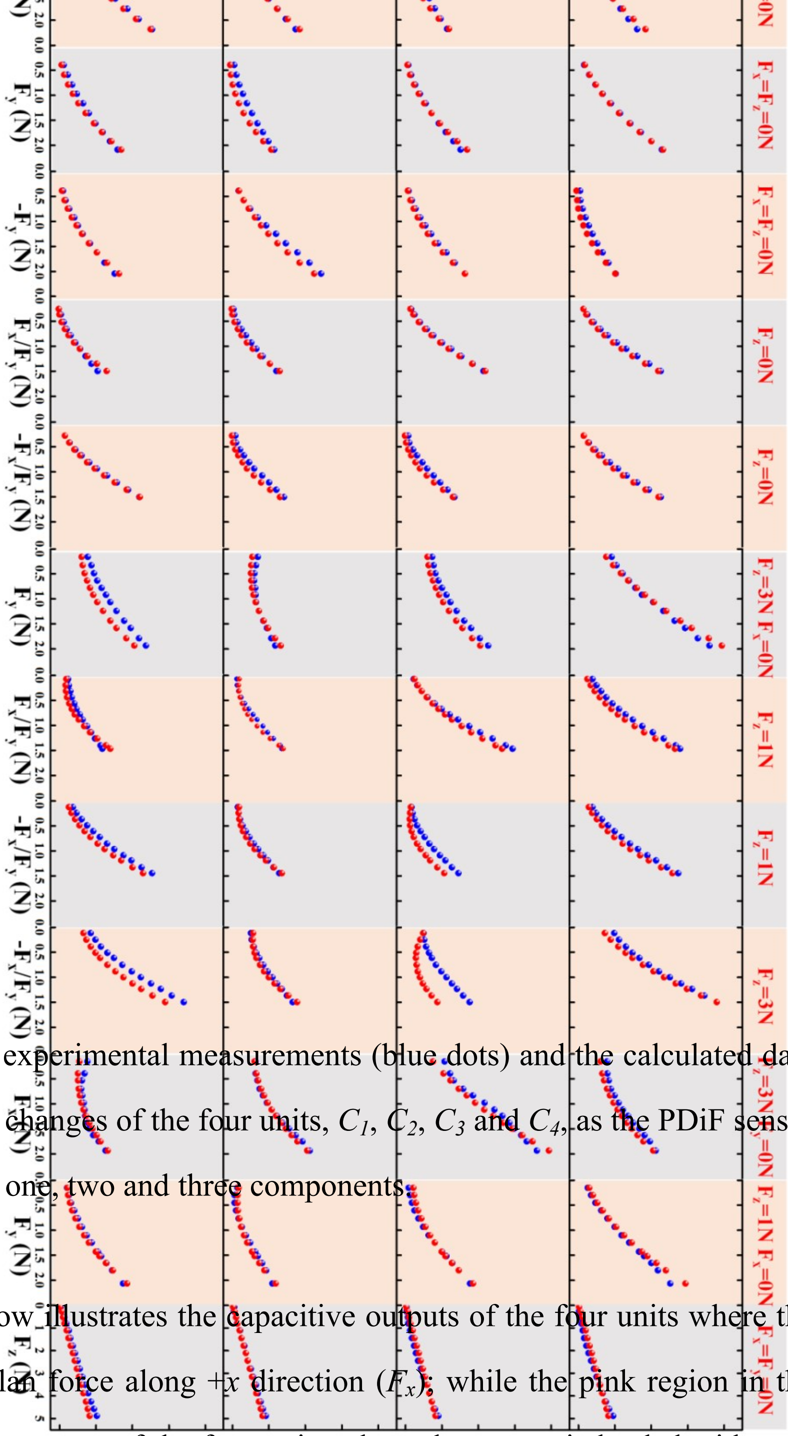

Figure S4 Comparison between the experimental measurements (blue dots) and the calculated data (red dots) of the relative capacitance changes of the four units, C_1, C_2, C_3 and C_4, as the PDiF sensor experiences with external loads with one, two and three components.

Notes: The grey region in the first row illustrates the capacitive outputs of the four units where the sensor is loaded with only one in-plan force along +*x* direction (F_x); while the pink region in the second row illustrates the capacitive outputs of the four units where the sensor is loaded with only one in-plan opposite force along -*x* direction (F_x). Interestingly, the pattern of the capacitance changes of C_2 in the first row is similar to that of C_4 in the second row, suggesting a high reproducibility of the units in force measurements. For more examples, the grey region in the 5th row gives the four capacitive increments as the PDiF sensor is taken an in-plane force with two components (F_x and F_y in equal magnitude); the grey region in the 9th row is the three component scenario where capacitances increase against an in-plane force in -*x* and +*y* directions complexed with a normal force (F_z) of 1N.

Mechanical and Dielectric Properties of the Porous Ecoflex Elastomer

Figure S5a illustrates the stress-strain curves of the solid and porous Ecoflex elastomers. The elastic modulus of porous elastomer is 33 kPa in the strain range of 0-10%, which is 5.5 times smaller than that of bulk elastomer. As the compression excess 10 %, a portion of pores can be closed, thus allowing the elastic modulus to increase to about 94 kPa. In addition, we have investigated the permittivity changes of the porous elastomer by sweeping the excitation frequency. Figure S5b shows the relative permittivity (ε_r) varies along with the excitation frequency ranging from 200 Hz to 1 M Hz under different levels of compression strain. The elastomer shows a similar trend on the dielectric decrease under different compression strains. The dielectric constant decreases by around 13.5% as the elastomer is compressed from 0 to 20 %.

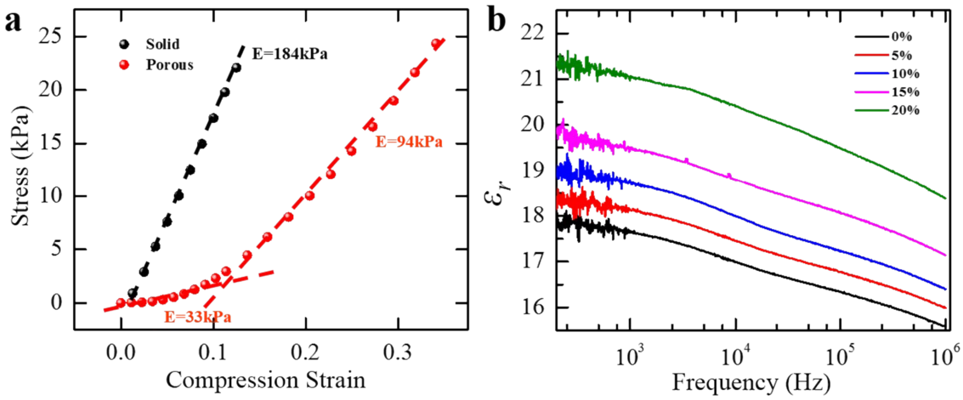

Figure S5 (a) Stress-strain curves of solid and porous Ecoflex elastomers. (b) Relative permittivity variations of the porous elastomer by sweeping the excitation frequencies under different levels of compression strains.

PDiF Sensor for Hand-Writing Recognitions

To extend the potential applications, our sensor has been used in hand-writing recognitions. More hand-writing letters are reconstructed by the PDiF-based "electronic pen". In Figure S6, both the uppercase and lowercase letters of "s", "u", "d" and "a" have been reconstructed (blue dots) by using the tracking algorithm to translate these capacitive values C_x and C_y into the corresponding coordinates. Meanwhile, the force intensity variations are presented by the dark dots.

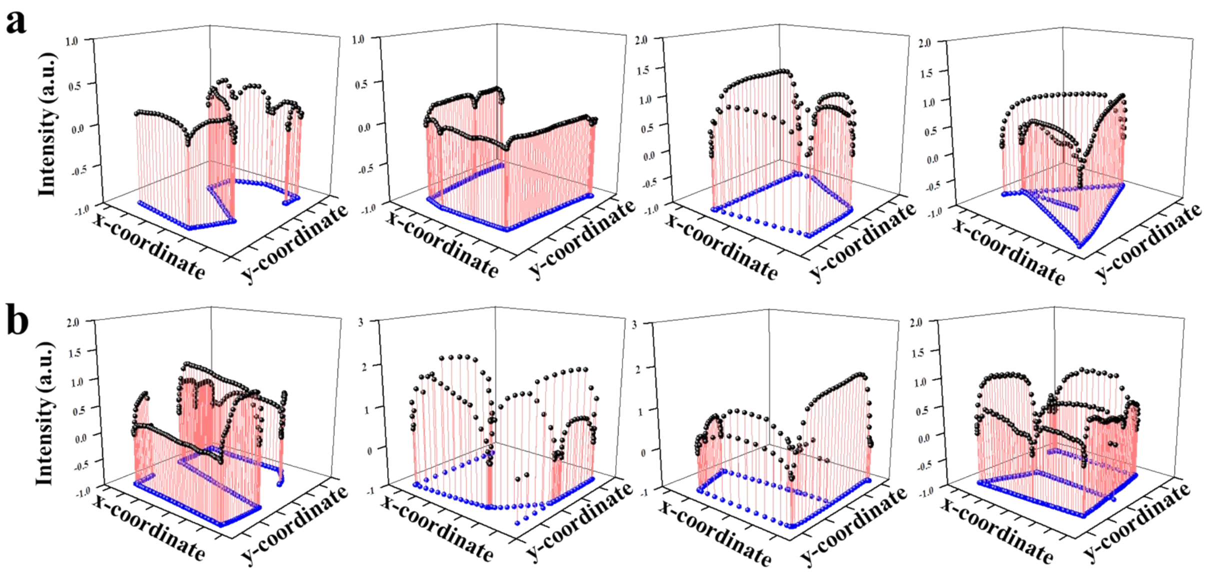

Figure S6 Demonstration of the PDiF sensor for hand-writing recognition. Writing and reconstructing in-plane upper-case (a) and lower-case (b) letters of "s", "u", "d" and "a" (blue dots), along with the corresponding variations of force intensity (dark dots).

Table S1 Comparison on the designs/features between the reports in the literature and our work

Ref	Sensing Mechanism	Sensing Materials	Features on Conductive /Dielectric Layer	Mode I Decoupling	Mode II Decoupling
H.K Lee et al, 2008 *J Microelectromech Syst*	Capacitive	Air	-	Nor or Tan separately	No
M.Y. Cheng et al, 2010 *Sensors*	Capacitive	Air	-	Nor or Tan separately	No
H.K. Lee et al, 2011 *J Micromech Microeng*	Capacitive	Air	-	Nor or Tan separately	No
J.A. Dobrzynska et al, 2013 *J. Micromech Microeng*	Capacitive	PDMS	Solid	Nor or Tan separately	No
L. Viry et al, 2014 *Adv Mater*	Capacitive	Air/solid fluorosilicone	Two layer stacked	Nor or Tan separately	No
G. Liang et al, 2015 *J Microelectromech S*	Capacitive	PDMS	Micro-texture on elastomer surface	Nor or Tan separately and combinatorially	Yes
Y. Song et al, 2018 *Micromachines*	Resistive	Conductive rubber	N-shape pillars	Nor or Tan separately and combinatorially	Yes
J. Yang et al, 2020 *Adv Funct Mater*	Capacitive	Liquid metal/Ecoflex	Porous	Nor	-
C. Lu et al, 2018 *Sens Actuator A: Phys*	Resistive	CNT/PDMS	Microstructured surface	Nor	-
C. Pang et al, 2012 *Nat Mater*	Resistive	Pt-coated /PUA	Interlocked nanohairs as bulk	Nor or Tan separately	No
C. Mu et al, 2018 *Adv Funct Mater*	Resistive	Graphene oxide/PDMS	Porous elastomer	Nor or Tan separately	No
Our Work	Capacitive	Li_2SO_4 /Ecoflex	Porous elastomer	Nor or Tan separately and combinatorially	Yes

Abbreviations:

PUA: polyurethane acrylate

PDMS: polydimethylsiloxane

CNT: carbon nanotube

Nor: normal

Tan: tangential