

Supplementary Information

High Sensitivity and Broad Linearity Range Pressure Sensor Based on Hierarchical In-Situ Filling Porous Structure

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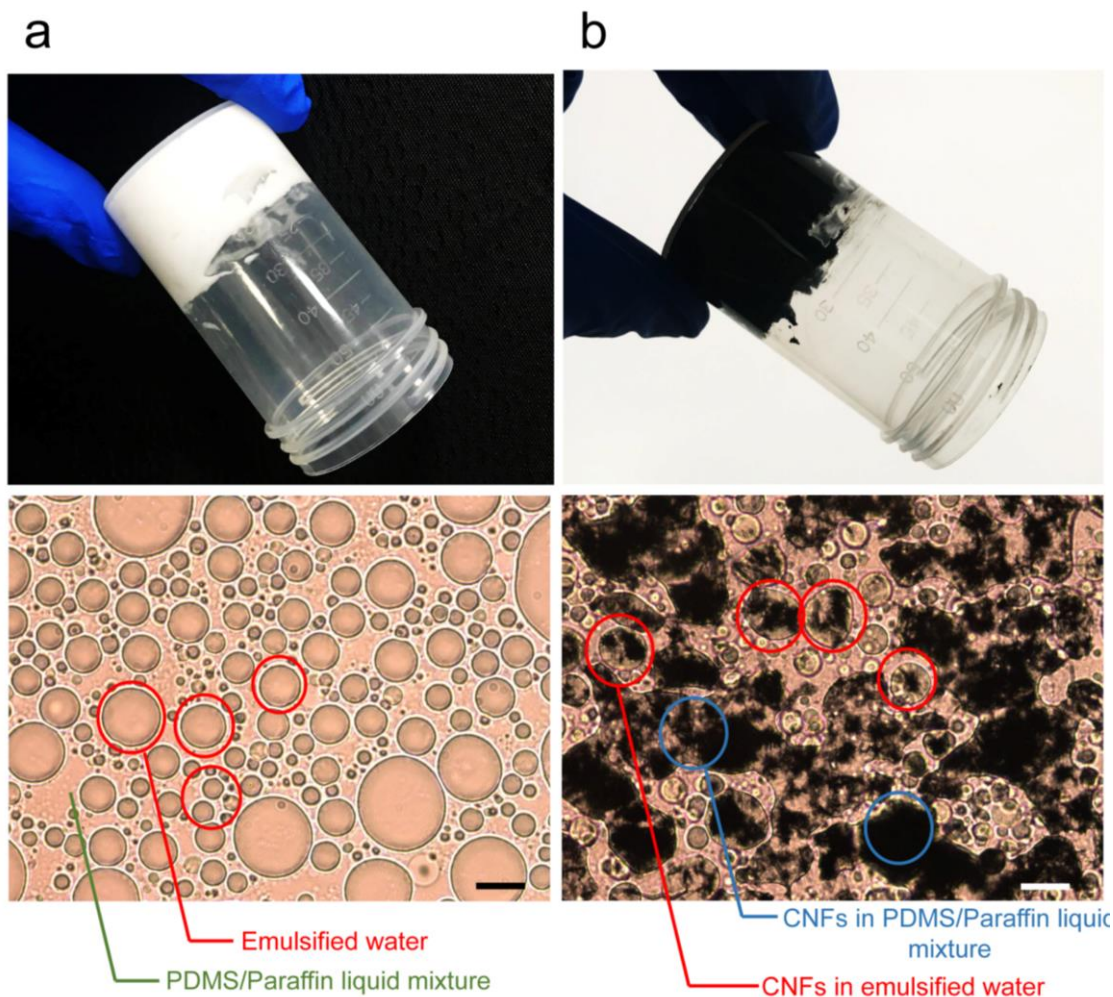
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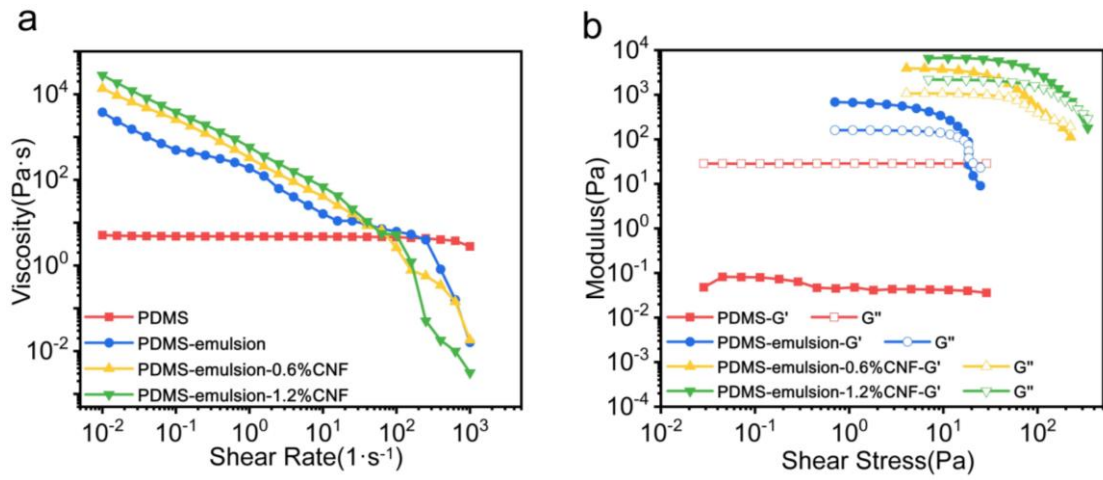
Supplementary Figure 1-27.

Supplementary Table 1-4.

Supplementary Video 1-5.



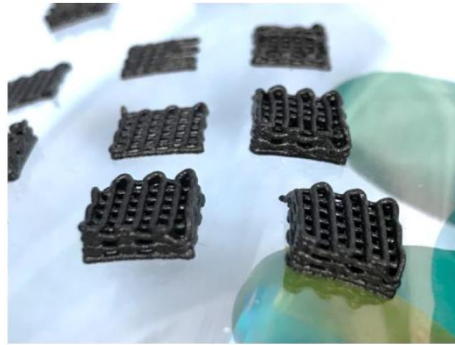
Supplementary Figure 1. Optical and microscope images of emulsion and CNFs/PDMS emulsion with porous structure. a. Optical image and microscope image of PDMS emulsion in an inverted vial (scale bar: 20 μm). **b.** Optical image and microscope image of CNFs/PDMS emulsion in an inverted vial (scale bar: 20 μm).



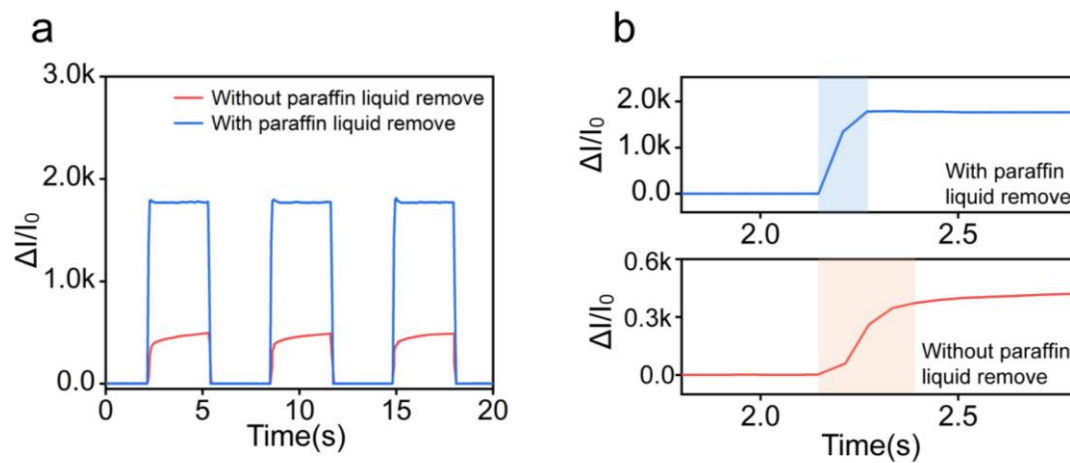
Supplementary Figure 2. Rheological performance of the CNFs/PDMS emulsion with different proportions of CNFs. a. The apparent viscosity as a function of shear rate. b. Storage modulus(G') and loss modulus(G'') as a function of shear stress.

a. The rheological measurements clearly suggest that the CNFs/PDMS emulsion inks are non-Newtonian fluids with shear-thinning behavior. Low viscosity at high shear ($10^2 \cdot s^{-1}$) rates suggest that inks can flow smoothly through the nozzle, and high viscosity at low shear ($10^{-2} \cdot s^{-1}$) rates indicate that inks could stack and preserve 3D shape integrity after leaving the nozzle. The viscosity is in proportion to the CNFs concentration. When more CNFs are added, the CNFs/PDMS emulsion will become more viscous, which is beneficial to preserve 3D shape integrity after printing.

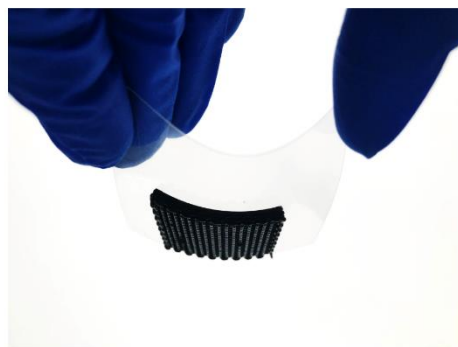
b. For the CNFs/PDMS emulsion inks, the storage modulus(G') is higher than the loss modulus(G'') at low shear stress, where the system resides in the linear viscoelastic region. This suggests that the inks are stiff with solid-like properties. When the shear stress increase to the yield point, the storage modulus is lower than the loss modulus, which indicated that the inks displayed liquid-like properties. The increased range of storage modulus at higher concentration CNFs contributes to the retention of the 3D-printed shape.



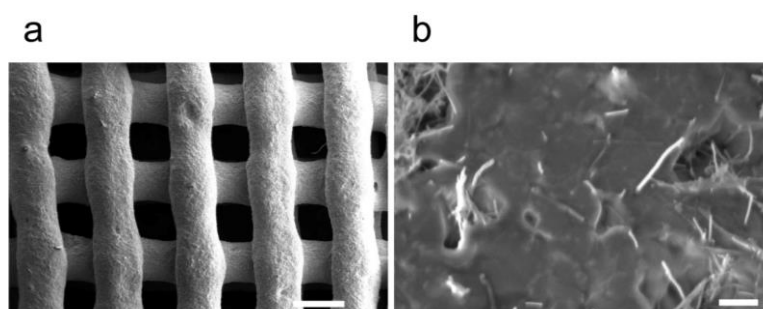
Supplementary Figure 3. Optical image of hierarchical in-situ filling porous structure with different lattice structures. Optical image of DIW continuously printed multiple and diverse structures to demonstrate the DIW technique is capable of mass and customized production.



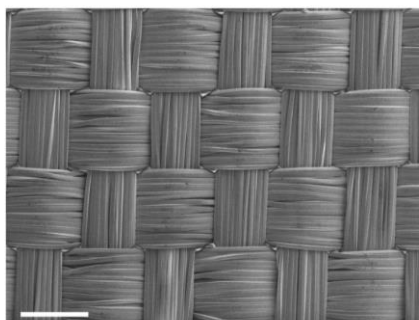
Supplementary Figure 4. a. sensing performance and b. response time comparison of sensors with and without liquid paraffin removal.



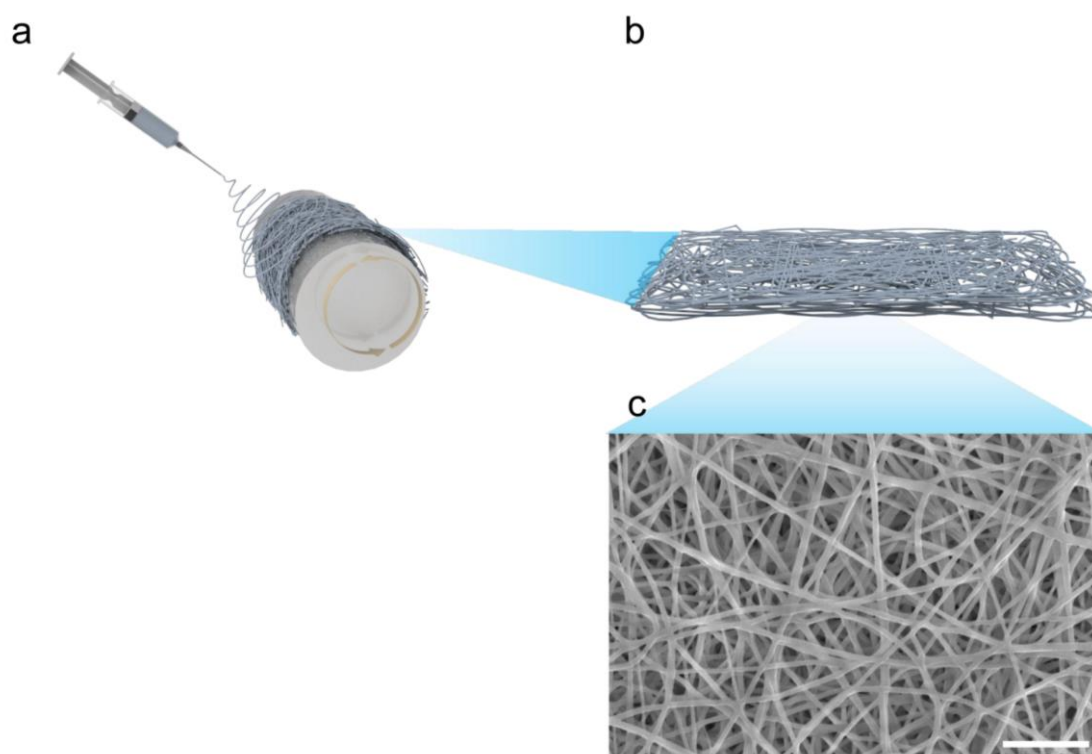
Supplementary Figure 5. Optical image of hierarchical in-situ filling porous sensing layer showing bending capacity.



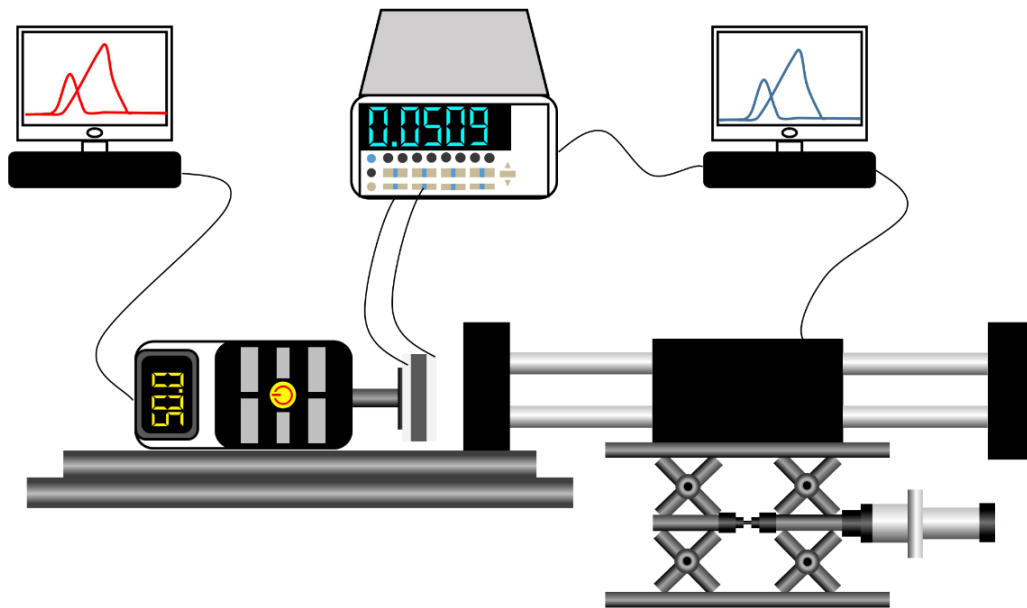
Supplementary Figure 6. SEM images. a. Top view of the printed hierarchical in-situ filling porous structure (scale bar: 500 μm). **b.** CNFs networks embedding in PDMS matrix (scale bar: 2 μm).



Supplementary Figure 7. SEM image of Ni conductive fabric tape (scale bar: 200 μm).

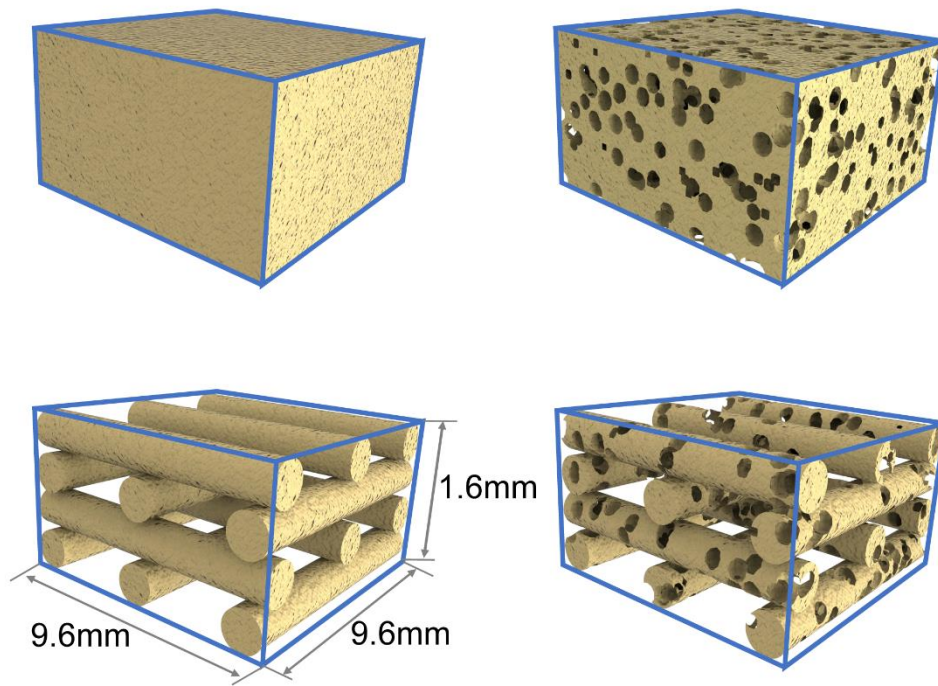


Supplementary Figure 8. Schematic of fabrication process of TPU film. a. Electrospinning process to fabricate **b.** TPU film. **c.** SEM image of TPU film (scale bar: 10 μm).

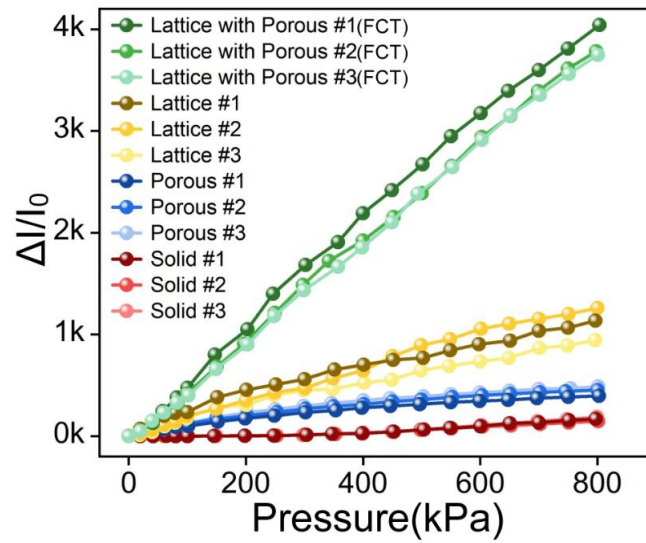


Supplementary Figure 9. Schematic of the pressure sensing measurement setup.

The force is provided and measured by a linear motor and a computer-controlled linear motor, respectively. The current is recorded in real-time using a LabVIEW-controlled digital source meter (Keithley 2611B), and the source-drain voltage was 0.1 V.



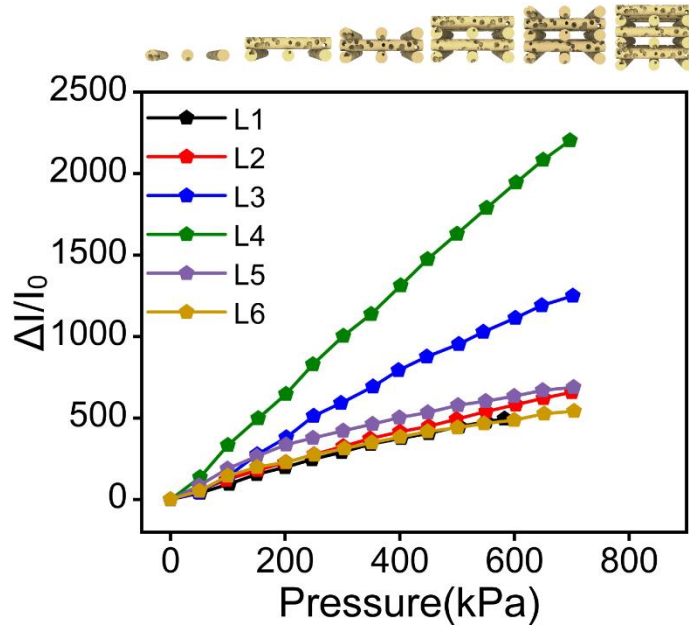
Supplementary Figure 10. Sample preparation with same extended contour volume for studies of the effect of geometric structure in current change and sensitivity.



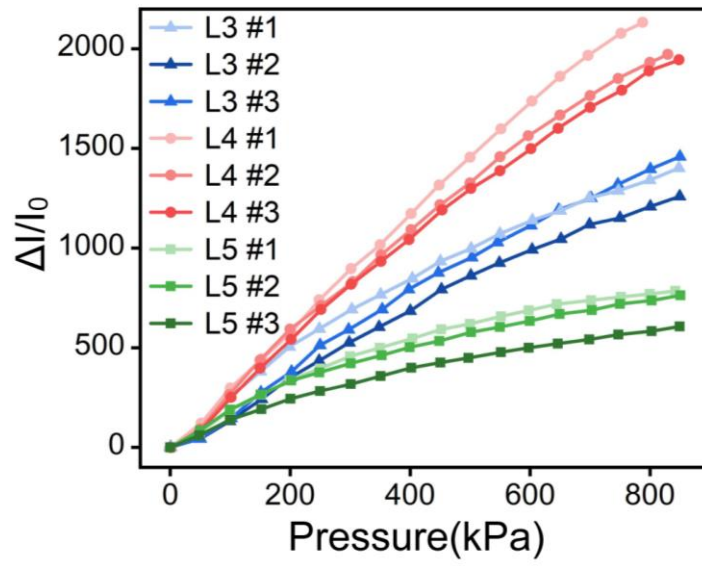
Supplementary Figure 11. The Current response of pressure sensor with different geometrical structures (Solid, Porous, Lattice, and Lattice with Porous (FCT)). Three samples were measured for each kind of geometrical structure.

	Lattice with porous(FCT)	Lattice	Porous	Solid
Sample 1	0.999	0.985	0.94	0.873
Sample 2	0.998	0.991	0.954	0.908
Sample 3	0.997	0.979	0.955	0.889

Supplementary Table 1. The linearity of pressure sensor with different geometrical structures.



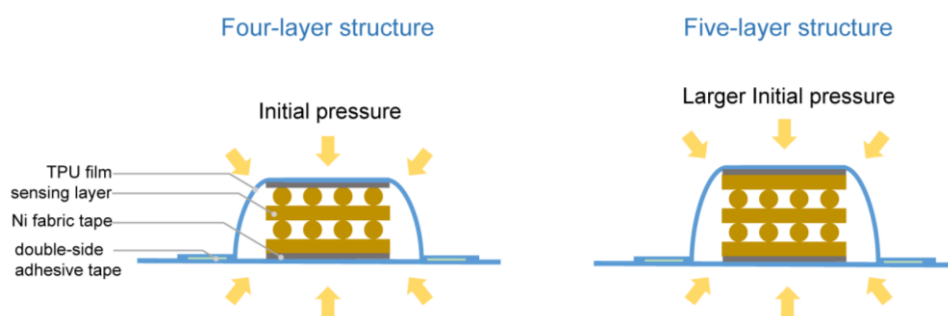
Supplementary Figure 12. Current response and sensitivity of HPSS with different numbers of sensing layer. Current response with printed different numbers of sensing layer. I_0 (5 layers) = 1.38 μA and I_0 (6 layers) = 1.4 μA are both large than then I_0 (4 layers) = 1 μA . Thus, the current change of five layers and six layers structures are lower than four layers structures.



Supplementary Figure 13. The comparison of the sensitivity of HPPS with 3, 4 and 5 layers of sensing layer by testing many samples.

Layer number	Sample	$I_0(\mu\text{A})$	$\Delta I(\text{mA})$	$\Delta I/I_0$
3 layer	1	0.984	1.38	1401
	2	0.923	1.34	1457
	3	0.977	1.23	1260
4 layer	1	1.12	2.38	2132
	2	1.01	1.99	1971
	3	1.10	2.14	1944
5 layer	1	2.88	2.26	785
	2	2.82	2.13	763
	3	3.62	2.19	606

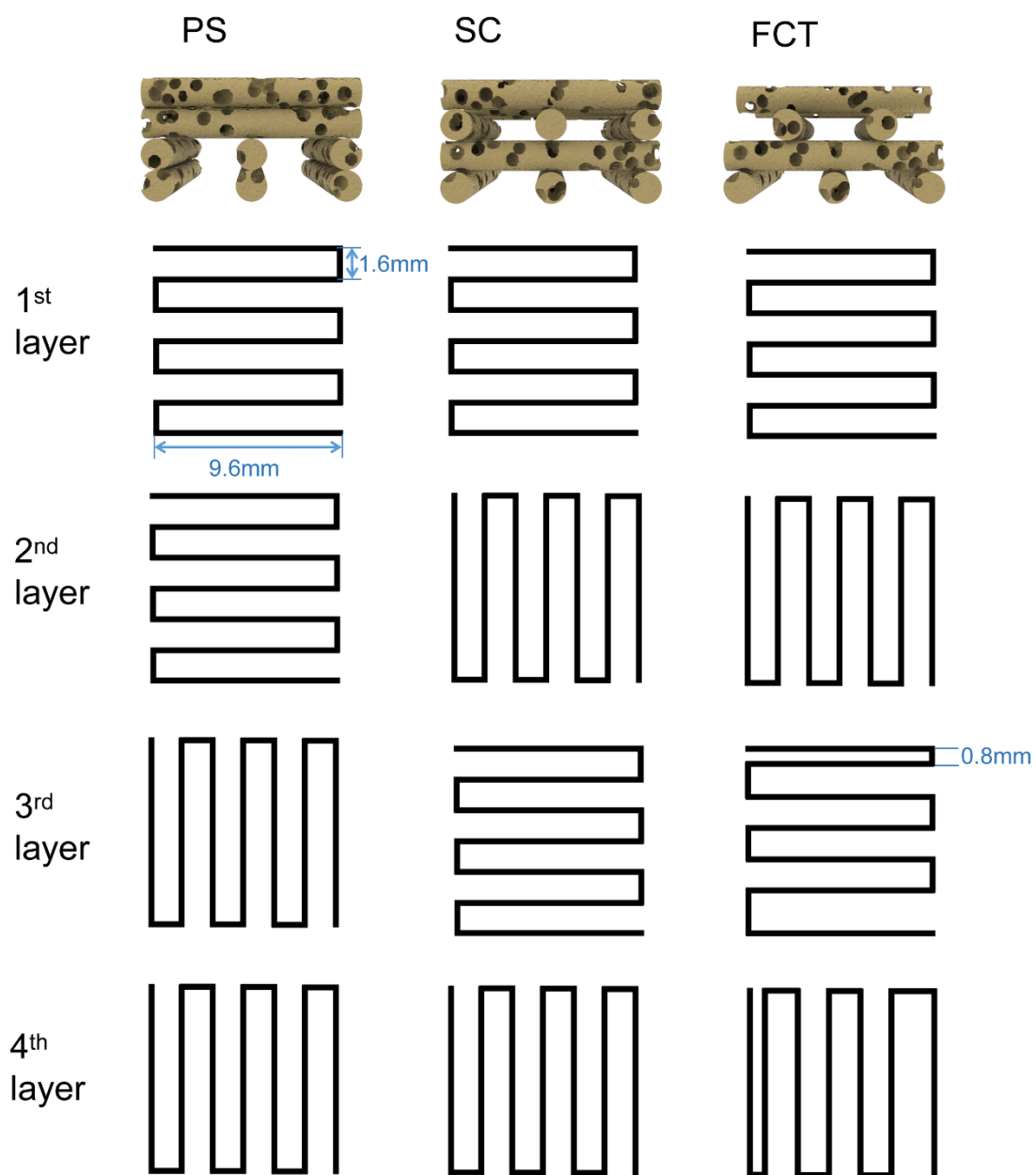
Supplementary Table 2. The comparison of the I_0 , ΔI , and $\Delta I/I_0$ of HPPS with 3, 4 and 5 layers of sensing layer by testing many samples. The I_0 of the four-layer structure is lower than the structure with more than four layers. When the pressure reaches 800 kPa, the final current of the four-layer structure and the structure with more than four layers are comparable. This results in $\Delta I/I_0$ of four-layer structure is larger than $\Delta I/I_0$ of the structure with the layer number of more than four layers structures. Compared with the one-, two-, and three-layer structures, the four-layer structure has more empty space between the lattice rods, resulting in higher deformability and more conductive pathway generation. The $\Delta I/I_0$ of four-layer structure is larger than $\Delta I/I_0$ of a structure with fewer than four layers.



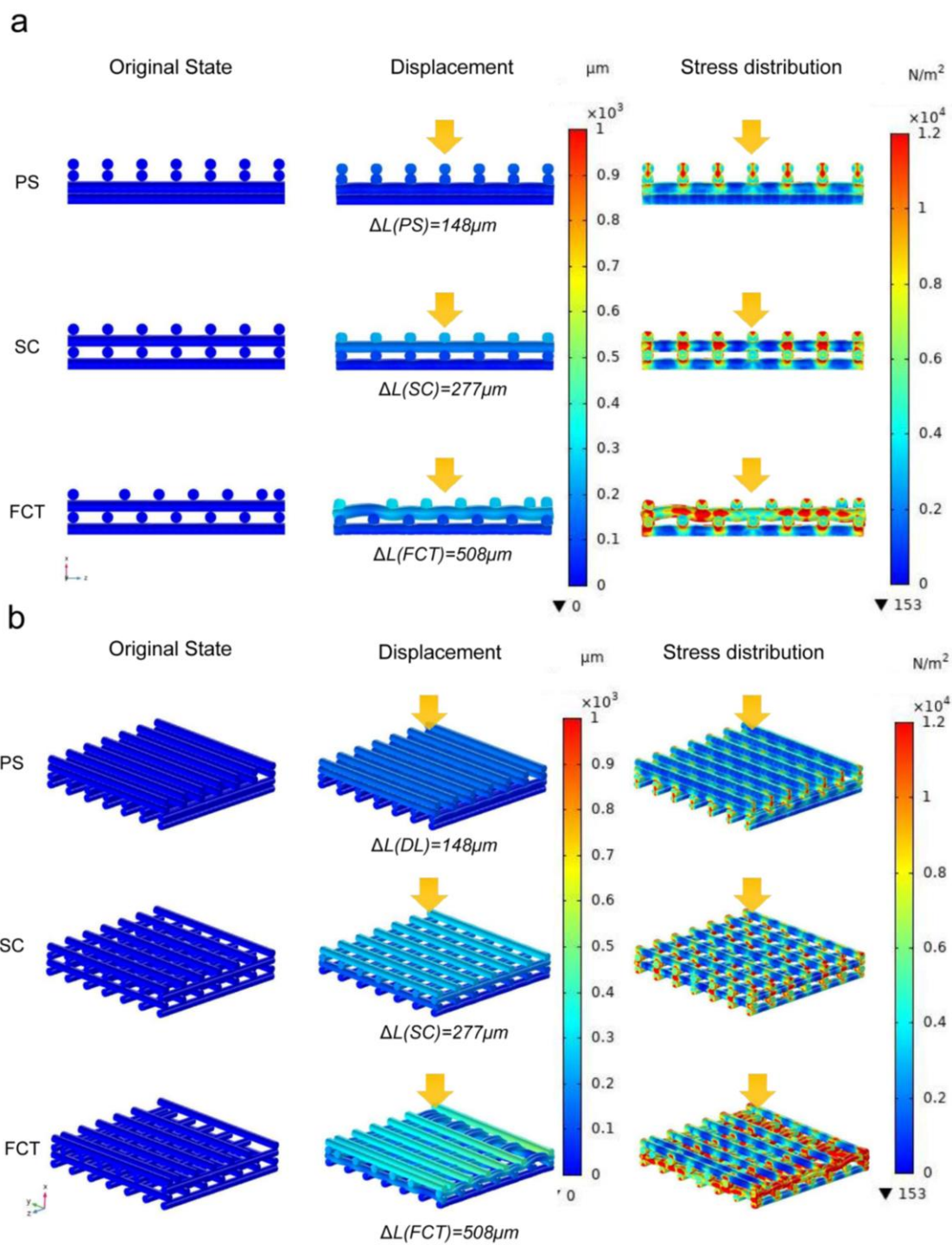
Supplementary Figure 14. Schematic cross-sectional view of the structure of the HPPS.

Samples	PDMS:water	m _(PDMS sponge) (g)	Porosity (%)
1	5:4	0.3169	34.86
2		0.3369	30.75
3		0.3269	32.80
4		0.3369	30.75
5		0.3169	34.86
6	5:7	0.2477	49.08
7		0.2558	47.42
8		0.2640	45.73
9		0.2730	43.88
10		0.2647	45.59
11	5:14	0.1968	59.54
12		0.2021	58.45
13		0.1987	59.15
14		0.1980	59.30
15		0.1933	60.26

Supplementary Table 3. The porosity of PDMS sponge under preparation conditions with different amounts of water.

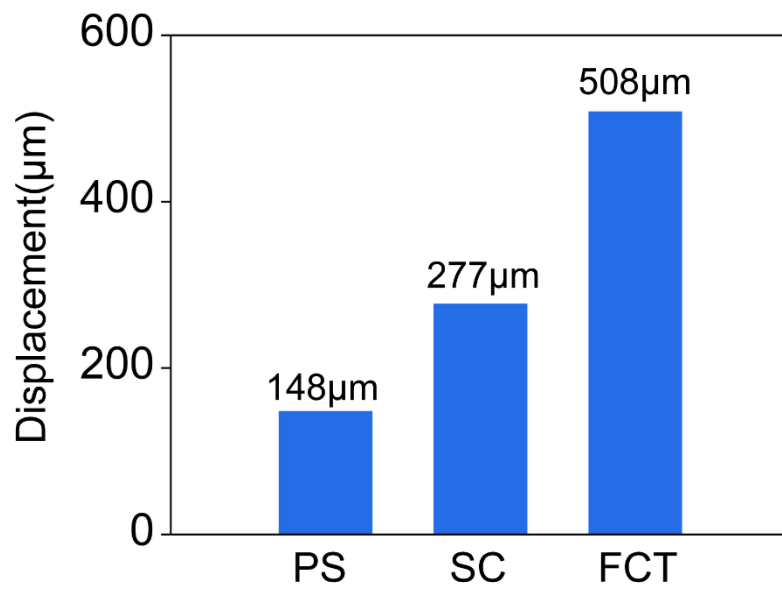


Supplementary Figure 15. Design diagram of lattice structure with different parameters.

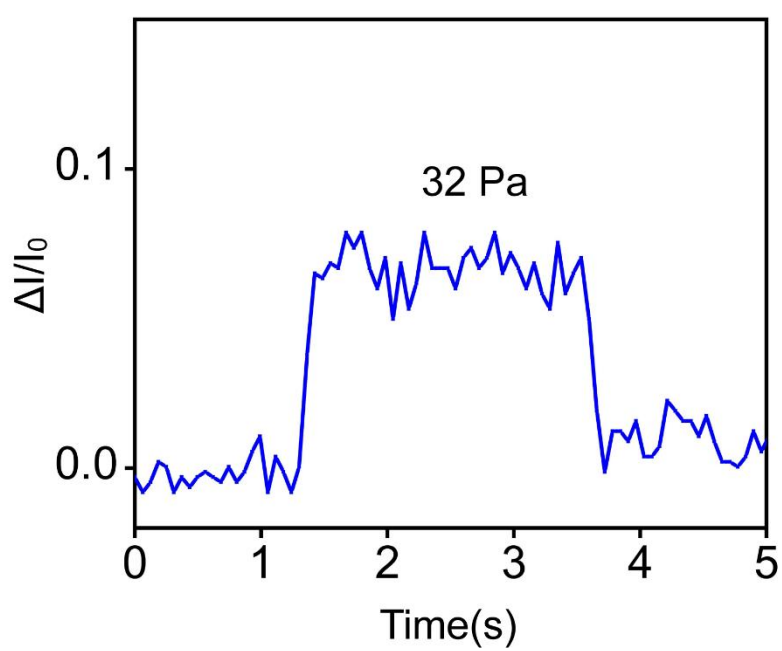


Supplementary Figure 16. Finite element analysis of PS, SC, FCT.

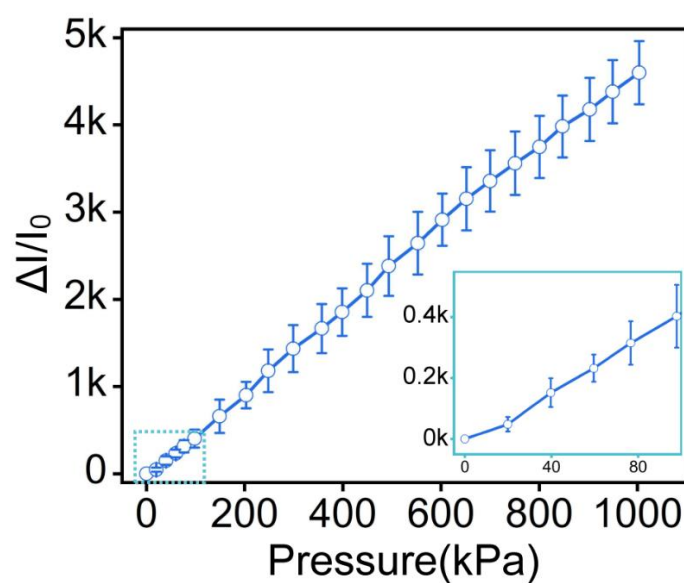
a. Cross-sectional view and **b.** Front view of finite element analysis of the displacement and local stress distribution of PS, SC, and FCT structures under the pressure of 1 MPa.



Supplementary Figure 17. Finite element analysis of PS, SC, and FCT. The comparison of the simulation displacement of PS, SC, and FCT structures.



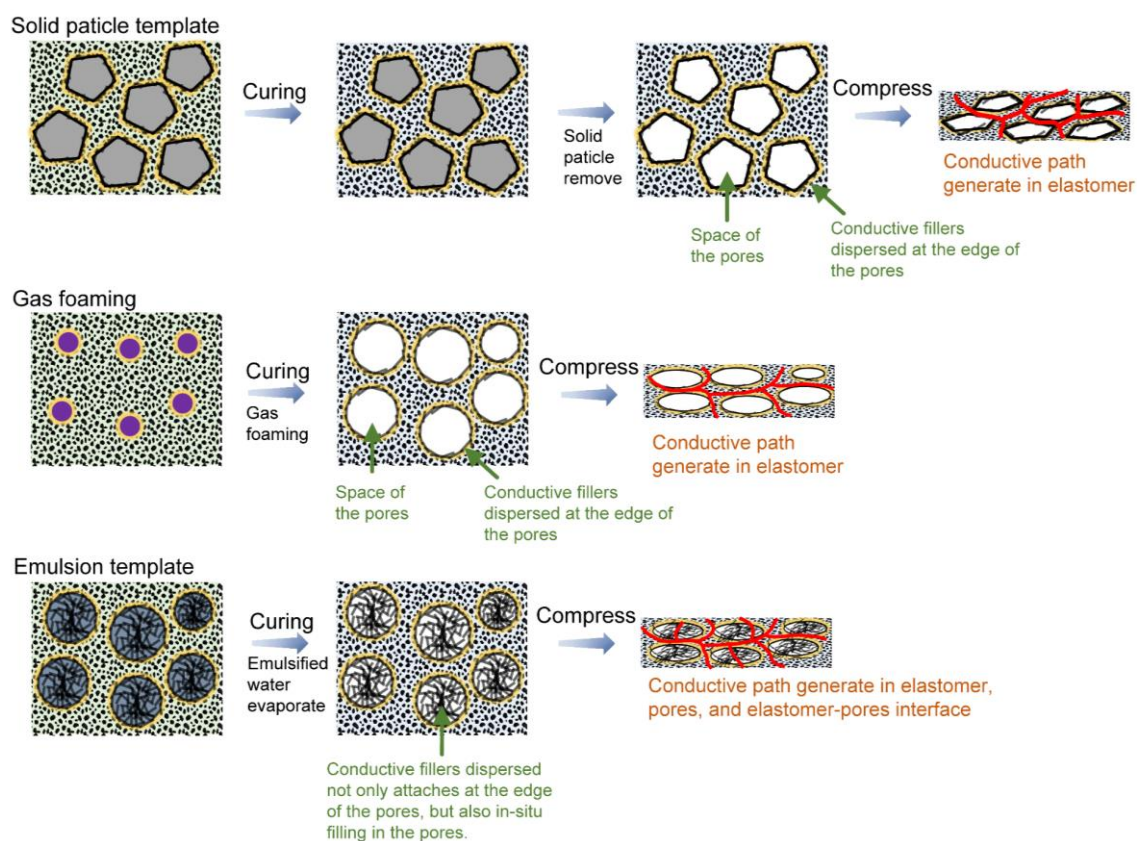
Supplementary Figure 18. The detection limit of HPPS. The relative current change under a subtle pressure of a half peanut.



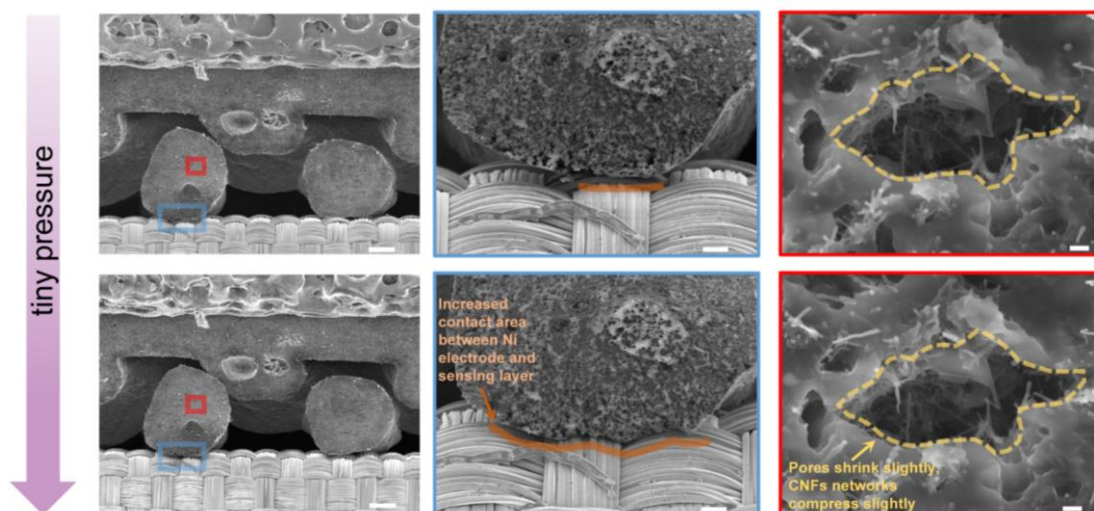
Supplementary Figure 19. Relative current change with error bars based on the optimized sensing layer under different pressures.

	Sensitivity(kPa⁻¹)	Linearity R²
Sample-1	5.01	0.994
Sample-2	4.96	0.995
Sample-3	4.75	0.998
Sample-4	4.74	0.998
Sample-5	5.03	0.997
Sample-6	5.08	0.998
Sample-7	4.86	0.998
Sample-8	5.04	0.993

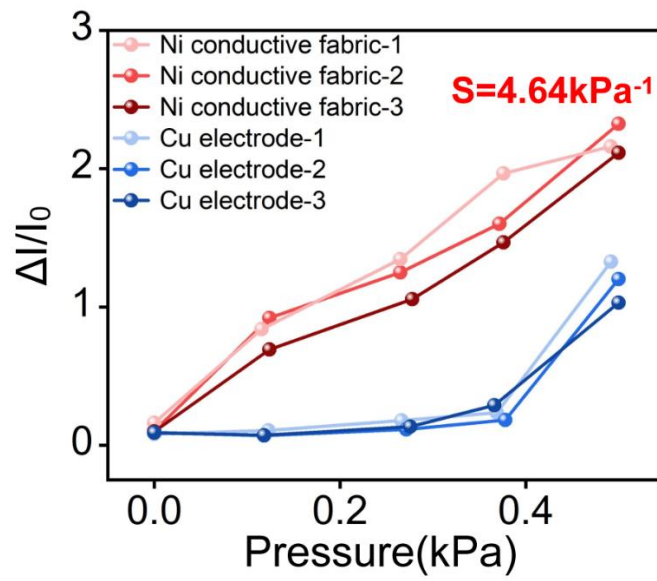
Supplementary Table 4. The sensitivity and linearity of eight different samples.



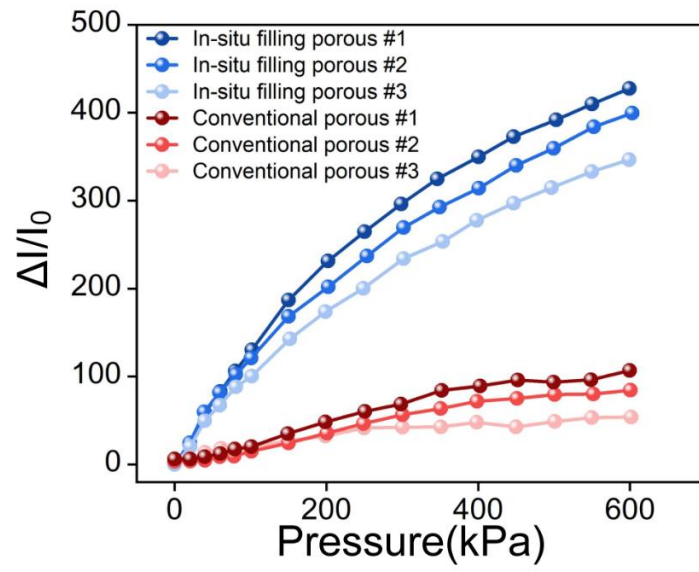
Supplementary Figure 20. Schematic diagram of porous structures prepared by different methods with different pressure-sensing mechanisms. Schematic illustration and comparison of porous structures formation by emulsion template method, sold particle template method and gas foaming method. The schematic illustration of pressure-sensing mechanisms of porous structures prepared by three different methods.



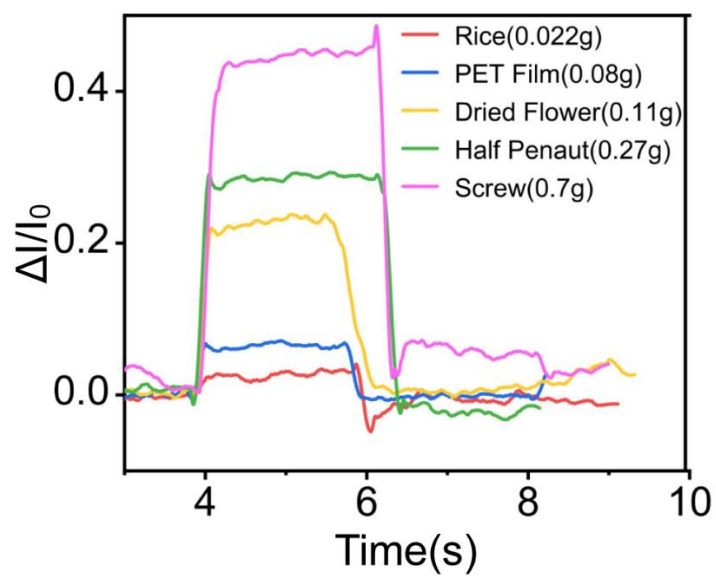
Supplementary Figure 21. In-situ SEM images of structural deformation of hierarchical in-situ filling porous structure under a tiny pressure (The scale bars of the three-column figures from left to right are: 200 μm , 50 μm , 2 μm).



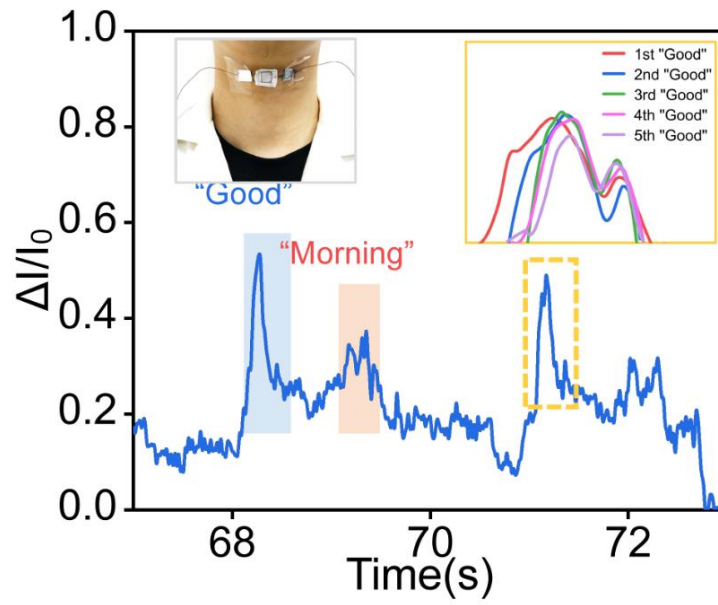
Supplementary Figure 22. The sensing performance of sensor fabricated using Ni conductive fabric and Cu electrodes at low pressure range.



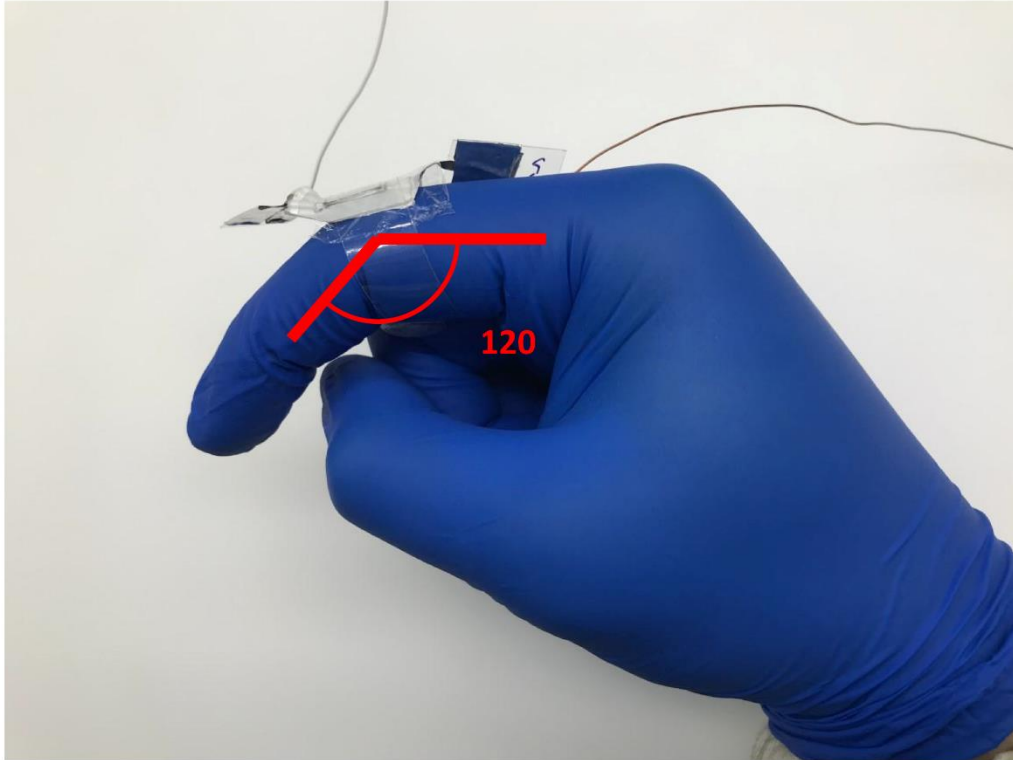
Supplementary Figure 23. The comparison of the sensitivity of in-situ filling porous sensor and conventional porous sensor.



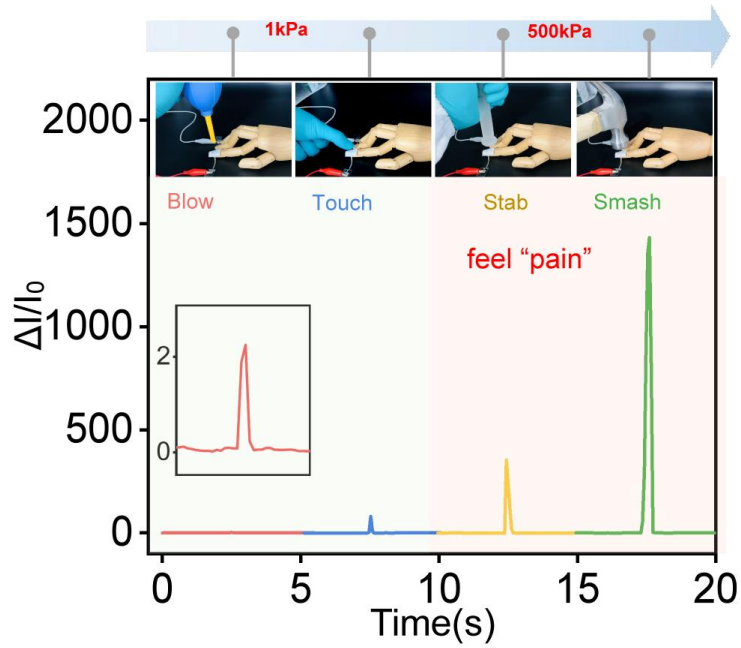
Supplementary Figure 24. Transient response to the loading/unloading light objects.



Supplementary Figure 25. Monitoring of throat vibration when pronounce ‘Good morning’. Repeatability of recognizing the same words ‘good’ for five times as shown in inset image.



Supplementary Figure 26. HPPS fixed on the index finger joint for monitoring finger bending angle. The angle of finger bending is defined as shown in the figure.



Supplementary Figure 27. Tactile recognition of different magnitudes of force, including blowing, touching, stabbing, and smashing. Sensors with a wide range can potentially be applied to tactile perception of human-machine interfaces. As shown in Supplementary Figure 27, the sensor is fixed on the finger of the hand model. The sensor can detect small pressures such as blowing, touching, and then larger pressures such as stabbing and smashing. This can potentially enable the robot hand to recognize the sensation of ‘pain’ based on the large current signal and make feedback actions to avoid injury by stab and smash. Furthermore, the distinct difference in current signal between stab and smash demonstrates that the sensor can distinguish different pressures even in the high-pressure range, and thus can potentially be used in pain severity grading.

Supplementary Video 1: Deformation of PS, SC, and FCT lattice structures under external pressure are simulated by FEA.

Supplementary Video 2: Relative current changes of the sensor fixed on the index finger joint for monitoring finger bending motions.

Supplementary Video 3: Relative current changes of the sensor fixed on the sole for monitoring human motions including walking, brisk walking, running and jumping.

Supplementary Video 4: Monitoring pressure change of a human weight under the preload of a car weight.

Supplementary Video 5: Monitoring pressure change of a box of water weight under the preload of a car weight.