Supporting Information

## Leveraging Temperature-Tunable, Scale-like Microstructure to Produce Multimodal, Supersensitive Sensors

Yanlong Tai<sup>1,2</sup>, Tushar Kanti Bera<sup>1</sup>, Zhenguo Yang<sup>2\*</sup>, Gilles Lubineau<sup>1\*</sup>

 <sup>1</sup> King Abdullah University of Science and Technology (KAUST), Division of Physical Science and Engineering, COHMAS Laboratory, Thuwal 23955-6900, Saudi Arabia
 <sup>2</sup> Fudan University, Department of Materials Science, Shanghai 200433, China.

\* Corresponding authors Email: <u>zgyang@fudan.edu.cn</u> (Zhenguo Yang), <u>gilles.lubineau@kaust.edu.sa</u> (Gilles, lubineau)

This PDF file includes:

1. Experimental videos.

Video-S1: Continuous monitoring of a human wrist and signals via a SSPF-based sensing device.

- Video-S2: The homogeneous and rapid response to temperature by a SSPF-2.
- Video-S3: Application of SSPF-2 for monitoring temperature and as a thermal switch (a fire alarming system).
- Video-S4: Application of SSPF-2 for monitoring light and as a light switch via a temperature mechanism.
- 2. Figure S1, Figure S2, and Figure S3: Further characterization of WSPFs and SSPFs.
- 3. Table S1, Table S2, Figure S4, and Figure S5: A summary of the recent literature about flexible pressure-sensing systems and temperature-sensing materials.
- 4. Figure S6: Response of SSPFs to light via a temperature mechanism.
- 5. Figure S7 and Figure S8: Analysis of the sensing mechanism of SSPFs.
- 6. Figure S9, Figure S10, Figure S11, Figure S12: Experimental Method.
- 7. References

AFM image of WSPFs with different drying temperatures



AFM image of SSPFs from the corresponding WSPFs



Variation in roughness between WSPFs and SSPFs



**Figure S1.** Atomic Force Microscope (AFM) images comparing the roughness between WSPFs and SSPFs.



**Figure S2.** The typical crack formation process from WSPF-2 to SSPF-2. a) WSPF-2; b) beginning of the cracking process; c) SSPF-2. All scale bars are 20  $\mu$ m.



**Figure S3**. The thermal deformation of a) WSPF-2, which bends due to its CTE value (PDMS: expansion, SWCNT: contraction) and b) SSPF-2, which has a fragmentation to its coating, as well as the corresponding illustration of deformations. The temperature was set at 100 °C.

Sample <sup>a</sup>	R <sub>0</sub>	Pressure/90%	∆R/90%	Sensitivity	Effective
	(10 <sup>3</sup> , ohm)	(break, kPa) <sup>ь</sup>	(10 <sup>6</sup> , ohm)	(break, kPa <sup>-1</sup> ) <sup>c</sup>	sensitivity (kPa <sup>-1</sup> ) <sup>d</sup>
SSPF-1	13.2	45/40.5	27.7/19.5	46.6	36.5
SSPF-2	2	63/56.7	35.1/24.6	278.6	216.9
SSPF-3	0.6	81/72.9	44.9/32.4	923.9	740.7

Table S1. A summary of the key responses of SSPFs to variation in pressure.

<sup>a</sup>: Sample size = a circle film with a diameter of 8 mm, and silver as electrode;

<sup>b</sup>: We define that the preferred detecting scope is 90 % of its breaking pressure load;

<sup>c</sup>: The sensitivity is defined as  $\Delta R/R_0$  to pressure;

<sup>d</sup>: The effective sensitivity is calculated from 90 % of its breaking pressure load with the corresponding variation in resistance.

Note that all data are the average values taken from three samples.



**Figure S4.** A summary of the recent literature with the key parameters for flexible pressuresensing systems.

Sample	R <sub>0</sub> (10 <sup>3</sup> , ohm)ª	Tem./90% (break, ℃) <sup>ь</sup>	R/90% (break, 10 <sup>6</sup> , ohm)	Thermal Index (break, 10 <sup>3</sup> , K) <sup>c</sup>	Effective Thermal Index (10 <sup>3</sup> . K <sup>-1</sup> ) <sup>d</sup>
SSPF-1	13.2	56.2/50.6	27.7/19.5	29.9	29.9
SSPF-2	2	73.2/65.9	35.1/24.6	20.4	20.4
SSPF-3	0.6	86.7/78	44.9/32.4	17.6	17.6

**Table S2.** A summary of the key parameters of SSPFs to variation in temperature.

<sup>a</sup>: Sample size = a circle film with a diameter of 8 mm and with silver as the electrode;

<sup>b</sup>: We define that the preferred detecting scope is 90 % of its breaking temperature;

<sup>c</sup>: The sensitivity is calculated according to Eq. 3;

<sup>d</sup>: The effective sensitivity is calculated from 90 % of its breaking temperature with the corresponding variation in resistance.

Note that all data are the average values taken from three samples.



## The representative materials

**Figure S5.** A summary of the recent literature with the key parameters for temperature-sensing materials.



**Figure S6.** The response of SSPF-2 to light. a) Relationship between temperature of SSPF-2 and distance from a light; b) the temperature distribution of the film with different distances from the light; c) light-monitoring performance; d) equivalent circuit and digital images for demonstrating the efficiency of SSPF-2 as a light switch. Note that the light is 100 W and the default distance is 10 cm.



**Figure S7.** SEM images illustrating the sensing mechanism of SSPFs. a) Deformation of SSPF without strain and with strain; the scale bar is 20  $\mu$ m. b) Macroscopic piezoresistive behavior of SSIs; the scale bar is 1  $\mu$ m. c) Microscopic piezoresistive behavior of SWCNT bundles; the scale bar is 500 nm.



**Figure S8.** SEM images and illustration for the evolution to WSPF transforming to SSPF and the deformation of SSPFs. All scale bars are 20  $\mu$ m.



**Figure S9.** a) Typical SWCNT ink (1 mg/ml); b) TEM of SWCNTs; c) Raman spectra with D, G, and G' band peaks, indicating the high purity of SWCNT. Note that the curve is generated directly from the ink sample, as shown by the inset image.



**Figure S10.** Scheme illustrations of the fabrication of SSPF-based pressure-sensing systems and the digital images of the typical devices a) as a unit and b) as an array.



**Figure S11**. Illustration and digital image of the setups used to test the sensitivity of films to mechanical pressure.



Figure S12. Digital image of the setups used to test for sensitivity to temperature.

## **Supporting Reference**

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