Supporting Information

Fibrous Strain Sensor with Ultra-sensitivity, Wide Sensing Range, and Large Linearity for Full-Range Detection of Human Motion

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1. Supplementary Table

Table S1 The performance comparison of strain sensors

Resistive-type strain sensors	Maximum	Limit of	Average gauge	Reference
	sensing range	detection	factor	
FPC strain sensor	150%	0.1%	80-350	This paper
Fragmentized graphene foam based sensor	70%	0.08%	29	S1
Carbonized cotton fabric based sensor	140%	0.02%	64	S2
graphene-based fiber with "compression	200%	0.2%	3.5	S3
spring" architecture				
Multilayered Au nanosheet films based	70%	a	36	S4
sensor				
ultrathin CPC layer-decorated PU yarn based	1%	0.1%	39	S5
sensor				
Parallel micro-cracked graphite film-based	50%	0.5%	500	S6
sensor				
Silver nanowire elastomer nanocomposite	70%	a	8	S7
based sensor				
Carbon nanotubes-Ecoflex nanocomposites	500%	a	2	S8
based sensor				
Aligned single-walled carbon nanotubes	300%	a	1	S9
based sensor				
ZnO nanowire/Polystyrene hybridized film	50%	1.5%	120	S10
based sensor				
Embedded 3D printing sensor	400%	a	5	S11
Graphene/natural rubber composite based	800%	a	32	S12
sensor				
Nanoscale cracked Pt/PET film	2%	0.5%	2000	S13
AuNWs/PANI film based sensor	100%	a	20.4-61.4	S14
AgNWs-PEDOT:PSS/PU nanocomposite	100%	1.5%	1.07-12.4	S15
sensor				
AuNWs/Latex rubber film based sensor	350%	0.5%	6.9-9.9	S16
CNT/PEDOT:PSS/PU nanohybrid based	100%	a	8.7-62.3	S17
sensor				
Graphene nanoplatelets coated yarn based	150%	a	2	S18
sensor				
Graphene-nanocellulose paper based sensor	100%	6%	8	S19
Graphene mesh fabric on stretchable tape	7.5%	a	20	S20
Fabricated by pencil drawn	0.6%	0.13%	536.6	S21
Laser scribed graphene on PDMS	10%	a	9.5	S22
Fish scale-like rGO/tape film	82%	0.1%	16.2	S23

			Clenching		"Victory" Gesture		
	Measured		Calculated Strain		Calculated Strain		
	GF	$\Delta R/R_0$	(%)	$\Delta K/K_0$	(%)		
X ₁₁	75	1.27	1.7	1.22	1.63		
X ₁₂	67	0.94	1.4	0.95	1.42		
X ₁₃	76.4	0.81	1.06	0.31	0.41		
X ₂₁	63	0.68	1.08	0.39	0.62		
X ₂₂	82	0.67	0.82	0.34	0.42		
X ₂₃	71	0.78	1.1	0.05	0.07		
X ₃₁	74	0.67	0.91	0.27	0.37		
X ₃₂	86	0.63	0.73	0.086	0.1		
X ₃₃	78	0.77	0.99	0.04	0.05		
Y ₁₁	69	2.5	3.6	1.66	2.4		
Y ₁₂	71	2.7	3.8	0.327	0.46		
Y ₁₃	70	2.59	3.7	0.006	0.009		
Y ₂₁	76	2.2	2.9	1.39	1.83		
Y ₂₂	67	2.08	3.1	0.35	0.52		
Y ₂₃	78	1.87	2.4	0.009	0.012		
Y ₃₁	76	1.6	2.1	1.14	1.5		
Y ₃₂	73	1.6	2.2	0.08	0.11		
Y ₃₃	83	1.33	1.6	0.002	0.003		

Table S2 The *GF* values of FPC strain sensors integrated into the sensing fabric and the resistance variation during clenching and "victory" gesture (X and Y are designed according to Fig. S12)

2. Supplementary Figures



Fig. S1 IR spectra of PEDOT:PSS, CNTs coated PU fiber, and PEDOT:PSS/CNTs coated PU fiber



Fig. S2 (a) Variation of current with strain of FPC strain sensors before and after the treat by methanol. (b) Relative resistance performance of FPC strain sensors before and after the treat by methanol.



Fig. S3 (a) SEM image of PEDOT:PSS/CNTs coated PU fiber core substrate; (b) magnified SEM image of PEDOT:PSS/CNTs coated PU fiber core substrate: the bottom of CNTs agglomerates are covered and fixed by the PEDOT:PSS layer.



Fig. S4 (a) Variation of current with strain of FPC strain sensors with 50% elongation of prestretching. (b) Relative resistance performance of FPC strain sensors with 75% elongation of pre-stretching.



Fig. S5 (a) Comparison of the morphologies of PU fiber cores with and without attachment of CNTs agglomerates. (b) SEM image of PU fiber cores with CNTs agglomerates. (c) SEM image of PU fiber cores without CNTs agglomerates. (d) Magnified SEM image of PU fiber cores with CNTs agglomerates. (e) Magnified SEM image of PU fiber cores without CNTs agglomerates.



Fig. S6 Fabrication process of the multi-pixel sensor array based on the FPC atrain sensors



Fig. S7 Mechanical behavior of (a) PU fiber; (b) Dragon Skin elastomer



Fig. S8 Variations of relative resistance of FPC strain sensor during the stretching-releasing process for 50% at the rate of 5% s⁻¹, 10% s⁻¹, 50% s⁻¹, respectively.



Fig. S9 Relative resistance response of the strain sensor to different strains after a stepped strain change.



Fig. S10 Mechanical stability of the PEDOT:PSS-CNTs/PU composite fiber upon cyclic stretching/releasing to 50% for 2000 cycles.



Fig. S11 Electromechanical behaviors of the fibrous strain sensor with only CNTs as the sensing layer. (a) $\Delta R/R_0$ - ϵ curve of the sensor recorded at a stretching rate of 1% sec⁻¹. (b)Variation in $\Delta R/R_0$ of the sensor under a 50% strain loading-unloading cycle.



Fig. S12 SEM image of the fibrous strain sensor without addition of CNTs at (a) 0% strain. (b) 30% strain. (c) 70% strain. (d) Magnified SEM image of the fibrous strain sensor without addition of CNTs at 70% strain. SEM image of the fibrous strain sensor without CNTs agglomerates at (e) 0% strain. (f) 30% strain. (g) 70% strain. (h) Magnified SEM image of the fibrous strain sensor without CNTs agglomerates at 70% strain sensor without CNTs agglomerates at 70% strain. (i) Schematic illustration of the fibrous strain sensor without CNTs agglomerates upon stretching. (j) $\Delta R/R_0 - \varepsilon$ curves of fibrous strain sensors with no CNTs and without CNTs agglomerates. The inset reproduces the curves within 0-50% strain range.



Fig. S13 FPC strain sensors integrated on the electronic fabric with 3×3 pixel array



Fig. S14 Cycling test of FPC strain sensor at a stretching/releasing rate of 1% s⁻¹; its relative resistance changes synchronously with strain.



Fig. S15 Plot of relative resistance as a function of bending degree. Inset: FPH strain sensor in bending testing.

3. Simulation of the sensing mechanism

Based on the microstructure variation of the FPC strain sensor, the resistance variation of the strain sensor was simplified into a network circuit model (Fig. 3d). In this model, total resistance of the sensor consists of the resistance of the PEDOT:PSS fragments wrapped on each PU fibers (R_1 , R_2 ... R_x , R_1 ', R_2 '... in Fig. 3d) and the contact resistance between PEDOT:PSS fragments and CNTs agglomerates (R_c in Fig. 3d). During uniaxial stretching, the resistance of every PEDOT:PSS fragments are approximated to be constant, and the increase in the resistance of the sensor is mainly attributed to the change of R_c . We simplified the change in R_c as the result of the stochastic break of the conductive path, and the proportion of the break is proportional to the strain.

By inputting the model into NI MultisimTM, the currents of the circuit with a specified proportional of the break can be calculated (spots in Fig. 3e). Based on our assumption that

the proportion of break is proportional to the strain, the variation of current with extensional strain can be simulated. Thus, the change of the $\Delta R/R_0$ with the strain (ε) is simulated, and fitted by using $\Delta R/R_0 - \varepsilon$ curves, as shown in Fig. 3e. Through changing the number of the conductive path (n in Fig. 3d), the $\Delta R/R_0 - \varepsilon$ curves of the FPC strain sensor with different PU fibers (m in Fig. 3d) can be obtained.

The simulated results show that the sensing architecture of FPC strain sensors will provide a good linear relationship between $\Delta R/R_0$ and ε at low strain, consistent well with the experimental results (Fig. 2d). However, this linear relationship will be terminated with the increase in the extensional strain, especially for the sensors with less PU fibers. In addition, the simulated curves show that *GF* (the slopes of $\Delta R/R_0 - \varepsilon$ curves) increases with the decrease of the PU fibers, consistent well with the experimental results in Fig. 2a. This could be attributed to the stronger influence of a quantitative break in the whole conductive network.

Supplementary References

S1 Y. R. Jeong, H. Park, S. W. Jin, S. Y. Hong, S. S. Lee, J. S. Ha, Adv. Funct. Mater. 2015, 25, 4228.

S2 M. Zhang, C. Wang, H. Wang, M. Jian, X. Hao, Y. Zhang, Adv. Funct. Mater. 2017, 27, 1604795.

S3 Y. Cheng, R. Wang, J. Sun, L. Gao, Adv. Mater. 2015, 27, 7365.

S4 G.-H. Lim, N.-E. Lee, B. Lim, J. Mater. Chem. C 2016, 4, 5642.

S5 X. Wu, Y. Han, X. Zhang, C. Lu, ACS Appl. Mater. Interfaces 2016, 8, 9936.

S6 M. Amjadi, M. Turan, C. P. Clementson, M. Sitti, *ACS Appl. Mater. Interfaces* **2016**, *8*, 5618.

- S7 M. Amjadi, A. Pichitpajongkit, S. Lee, S. Ryu, I. Park, ACS Nano 2014, 8, 5154.
- S8 M. Amjadi, Y. J. Yoon, I. Park, Nanotechnology 2015, 26, 375501.
- S9 T. Yamada, Y. Hayamizu, Y. Yamamoto, Y. Yomogida, A. Izadi-Najafabadi, D. N. Futaba, K. Hata, *Nat. Nanotechnol.* **2011**, *6*, 296.
- S10 X. Xiao, L. Yuan, J. Zhong, T. Ding, Y. Liu, Z. Cai, Y. Rong, H. Han, J. Zhou, Z. L.
 Wang, *Adv. Mater.* 2011, *23*, 5440.
- S11 J. T. Muth, D. M. Vogt, R. L. Truby, Y. Mengüç, D. B. Kolesky, R. J. Wood, J. A.
 Lewis, *Adv. Mater.* 2014, *26*, 6307.
- S12 C. S. Boland, U. Khan, C. Backes, A. O'Neill, J. McCauley, S. Duane, R. Shanker,Y. Liu, I. Jurewicz, A. B. Dalton, *ACS Nano* 2014, *8*, 8819.
- S13 D. Kang, P. V. Pikhitsa, Y. W. Choi, C. Lee, S. S. Shin, L. Piao, B. Park, K.-Y. Suh, T.i. Kim, M. Choi, *Nature* 2014, *516*, 222.
- S14 S. Gong, D. T. Lai, Y. Wang, L. W. Yap, K. J. Si, Q. Shi, N. N. Jason, T. Sridhar, H. Uddin, W. Cheng, ACS Appl. Mater. Interfaces 2015, 7, 19700.
- S15 J.-H. Lee, B.-U. Hwang, T. Q. Trung, E. Roh, D.-I. Kim, S.-W. Kim, N.-E. Lee, ACS
 Nano 2015, 9, 8801.
- S16 S. Gong, D. T. H. Lai, B. Su, K. J. Si, Z. Ma, L. W. Yap, P. Guo, W. Cheng, *Adv. Electron. Mater.* **2015**, *1*, 1400063.
- S17 E. Roh, B.-U. Hwang, D. Kim, B.-Y. Kim, N.-E. Lee, ACS Nano 2015, 9, 6252.
- S18 J. J. Park, W. J. Hyun, S. C. Mun, Y. T. Park, O. O. Park, ACS Appl. Mater. Interfaces 2015, 7, 6317.
- S19 C. Yan, J. Wang, W. Kang, M. Cui, X. Wang, C. Y. Foo, K. J. Chee, P. S. Lee, *Adv. Mater.* 2014, *26*, 2022.
- S20 Q. Liu, M. Zhang, L. Huang, Y. Li, J. Chen, C. Li, G. Shi, ACS Nano 2015, 9, 12320.
- S21 X. Liao, Q. Liao, X. Yan, Q. Liang, H. Si, M. Li, H. Wu, S. Cao, Y. Zhang, *Adv. Funct. Mater.* 2015, 25, 2395.

- S22 H. Tian, Y. Shu, Y.-L. Cui, W.-T. Mi, Y. Yang, D. Xie, T.-L. Ren, *Nanoscale* 2014, 6, 699.
- S23 Q. Liu, J. Chen, Y. Li, G. Shi, ACS Nano 2016, 10, 7901.