Sea urchin-like microstructures pressure sensors with an ultra-broad

range and high sensitivity

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Supplementary Figures



Supplementary Figures 1. XRD patterns. **a** the different mass ratios of Fe_2O_3/C (4:1,3:1,2:1,1:1,1:2), **b** the different mass ratios of $Fe_2O_3/C@SnO_2(3:1:1,3:1:4,3:1:8)$



Supplementary Figure 2. Fe₂O₃/C and Fe₂O₃/C@SnO₂ microparticle morphology. The SEM images of the different mass ratios of Fe₂O₃/C (\mathbf{a} 4:1, \mathbf{b} 2:1, \mathbf{c} 1:1, \mathbf{d} 1:2), the different mass ratios of Fe₂O₃/C@SnO₂ (\mathbf{e} 3:1:1, \mathbf{f} 3:1:8, \mathbf{g} 3:1:4), \mathbf{g} and \mathbf{h} the mass ratio of Fe₂O₃, C, and SnO₂ 3:1:4 after loading-unloading for 3500 cycles.**i.** High resolution TEM(HRTEM) images of the mass ratio of Fe₂O₃, C and SnO₂ 3:1:4.



Supplementary Figure 3. SEM images. **a** and **b** SEM images of melamine sponge, **c** and **d** SEM images of Fe₂O₃/C@SnO₂/ melamine sponge.



Supplementary Figure 4. Pressure-sensing characterizations. **a** Relative current change for the applied pressure, where the curves represent the different mass ratios of Fe₂O₃/C (4:1, 3:1, 2:1, 1:1 and 1:2), **b** The different mass ratios of Fe₂O₃/C@SnO₂ (3:1:1, 3:1:4 and 3:1:8), **c** Fe₂O₃ and SnO₂, **d** C and the mass of SnO₂@C 4:1.



Supplementary Figure 5. Pressure-sensing characterizations. **a** The sensitivity of Fe₂O₃, SnO₂, and Fe₂O₃/SnO₂ pressure sensor. **b** The sensitivity of Fe₂O₃/C@ Fe₂O₃ (3:1:4) and Fe₂O₃/C@ SnO₂ (3:1:4) pressure sensor. **c** The sensitivity of Fe₂O₃/C@ Fe₂O₃ with the mass of ratio of 3:1:4 based sensors.



Supplementary Figure 6. SEM images of Fe₂O₃/C@ Fe₂O₃ (3:1:4)



Supplementary Figure 7. Sensing performance under different relative humidity. Sensitivities of $Fe_2O_3/C@SnO_2$ (3:1:4) pressure sensor under the RH of a 73%, b 85% and c 95% at room temperature.



Supplementary Figure 8. Sensing performance under different areas and thickness. The current ratio variation with pressure of the Fe₂O₃/C@SnO₂(3:1:4) sponge **a-c** with a sizes of 15×20 mm, 19×19 mm and 25×25 mm, **d-f** thickness of 2 mm, 4 mm and 8 mm, respectively.



Supplementary Figure 9. Detection of micro pressure under loading pressures. **a** 2.8 kPa and **b** 50 kPa.

Supplementary faster Sensitivity summary of pressure sensor in this work				
Materials	Sensitivity(kPa ⁻¹) (0-10 kPa)	Sensitivity (kPa ⁻¹) (10-50 kPa)	Sensitivity (kPa ⁻¹) (50-150 kPa)	
Fe ₂ O ₃	3	3	2	
SnO_2	1	1	0.60	
С	15	0.88	0.20	
Fe_2O_3/C (4:1)	88	7	2.30	
Fe_2O_3/C (3:1)	203	44	6	
Fe_2O_3/C (2:1)	76	9	3.90	
Fe_2O_3/C (1:1)	47	8	1.27	
Fe_2O_3/C (1:2)	28	3	1.02	
SnO ₂ /C (4:1)	13	2.4	0.7	
$Fe_2O_3/C@SnO_2(3:1:1)$	420	80	12	
$Fe_2O_3/C@SnO_2(3:1:4)$	680	98	35	
$Fe_2O_3/C@SnO_2(3:1:8)$	314	65	11	

Supplementary Table 1 Sensitivity summary of pressure sensor in this work

Materials	Nanostructure	Sensitivity (kPa ⁻¹)	Reference
ZnO/PDMS	Sea urchin-like	75 - 121 (0 - 200 Pa)	1
Au/Ag/PU	Sea urchin-like	2.46 (0 - 1 kPa) 0.52 (1 -8.2 kPa)	2
C/PDMS	Sea urchin-like	263 at 1 Pa	3
Mxene/Reduced Graphene Oxide Aerogel	Naosheets	0.55 (23 - 982 Pa) 3.81 (982Pa - 10 kPa) 2.52 (10 - 30 kPa)	4
Carbon Black@Polyurethane Sponge	Nanosheets	0.068 (0-2 kPa) 0.023 (2-10 kPa) 0.036 (10-16 kPa)	5
MXene/Sponge	Nanosheets	147 (0-5 kPa) 442 (5-20 kPa)	6
Graphene/Eco-flex	Triode-mimicking	4.68 (0-150 kPa) 11.09 (150-200 kPa)	7
CNT/cotton textile	Nanowires	14.4 (0-3.5 kPa) 7 (3.5-15 kPa)	8
MXene/tissue paper	Nanosheets	0.55 (0-3 kPa) 3.81 (3-10 kPa)	9
Fe ₂ O ₃ /C@SnO ₂ /Spo nge	Sea urchin-like	680 (0-10 kPa) 98 (10-50 kPa) 35 (5-150 kPa)	In this work
E _C E _F E _V			
	n-Fe ₂ O ₃	n-SnO ₂	

Supplementary Table 2 Sensitivity summary of pressure sensor in reference

Supplementary Figure 10. Schematic illustration of formed Fe_2O_3 -SnO₂ n-n heterojunction. E_{CB} : conduction band edge energy, E_{VB} : valence band edge energy, E_{gap} : band gap energy, E_F : fermi energy.



Supplementary Figure 11. Change of current with pressure increases for Fe₂O₃/C@Sb₂O₃.



Supplementary Figure 12. The schematic pressure-sensing models of the sponge.

Reference

- 1. Yin, B., Liu, X., Gao, H., Fu, T., & Yao, J. Bioinspired and bristled microparticles for ultrasensitive pressure and strain sensors. *Nat. Commun.* 9, 1-8 (2018).
- 2. Lee, D. et al. Highly sensitive, transparent, and durable pressure sensors based on

sea-urchin shaped metal nanoparticles. Adv. Mater. 28, 9364-9369 (2016).

- 3. Shi, L. et al. Quantum effect-based flexible and transparent pressure sensors with ultrahigh sensitivity and sensing density. *Nat. Commun.* **11**,1-9 (2020).
- 4. Ma, Y. et al. 3D synergistical MXene/reduced graphen oxide aerogel for a piezoresistive sensor. *ACS Nano*, **12**, 3209-3216 (2018).
- 5. Wu, X. et al. Large-Area Compliant, Low-Cost, and Versatile Pressure-Sensing Platform Based on Microcrack-Designed Carbon Black@Polyurethane Sponge for Human-Machine Interfacing. *Adv. Funct. Mater.* **26**, 6246-6256 (2016).
- 6. Yang, Y. et al. 3D hybrid porous Mxene-sponge network and its application in piezoresistive sensor. *Nano Energy*. **50**, 79-87 (2018).
- 7. Wu, Q. et al. Triode-mimicking graphene pressure sensor with positive resistance variation for physiology and motion monitoring. *ACS Nano.* **14**, 10104-10110 (2020).
- 8. Liu, M. et al. Large-Area All-Textile Pressure Sensors for Monitoring Human Motion and Physiological Signals. *Adv. Mater.* **29**,701-709 (2017).
- Ying, et al. A Wearable Transient Pressure Sensor Made with MXene Nanosheets for Sensitive Broad-Range Human-Machine Interfacing. *Nano lett.* 19, 1143-1150 (2019).