# Electronic supplementary information

## Contact resistance based tactile sensor using covalently cross-linked

### graphene aerogels

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**Figure S1**: (a) Actual pictures of the materials obtained after every step of synthesizing 3D graphene aerogels (scale bar: 1 cm). (b) actual photograph of home-made high temperature vacuum furnace.



**Figure S2**: SEM micrograph of non-annealed graphene aerogel (a) uncompressed state, (b) compressed state, (c-d) actual photograph of non-annealed samples in uncompressed and compressed state.



Figure S3: XPS survey scans of a 3D graphene aerogel before and after annealing.

Sample	Etch (sec)	C (at.%)	O (at.%)
<b>Before annealing GA</b>	0	88.2	11.8
	30	88.7	11.3
After annealing GA	0	98.4	1.6
	30	98.3	1.7

Table S1: XPS compositional analysis of a 3D graphene aerogel before and after the annealing.

Table S2: Deconvolution peak analysis of C 1s peaks in 3D graphene aerogel before and after annealing.

Sample	Peaks	C sp <sup>2</sup>	C sp <sup>3</sup>	C-0	<b>O-C=O</b>	π- π*
Before	Position (eV)	284.4	285.0	286.0	288.5	291.5
annealing	FWHM (eV)	0.7	0.9	1.6	3.3	5.6
GA	Area %	53.7	11.4	13.6	13.3	8
After	Position (eV)	284.4	285.0	286.0	287.5	291.2
annealing	FWHM (eV)	0.6	0.9	1.9	3.7	5.7
GA	Area %	70.5	8.9	6.1	6.1	8.4

Table S3: Deconvolution peak analysis of O 1s peak in 3D graphene aerogel before and after annealing.

Sample	Peaks	C-0	C=0	<b>O-C=O</b>
Before	Position (eV)	533.1	531.7	530.7
annealing GA	FWHM (eV)	1.8	1.8	1.8
	Area %	50.8	28	21.2
After	Position (eV)	533.0	531.9	-
annealing GA	FWHM (eV)	1.8	1.8	-
	Area %	63.4	36.6	-



Figure S4: Raman spectroscopy and XPS analysis of graphene aerogels annealed at different temperatures in vacuum.



**Figure S5**: The stress-strain curve of graphene aerogel in (a) compression and (b) tensile. (c) actual photographs of non-annealead and annealead graphene aerogel before and after loading (scale bar: 5 mm).



**Figure S6**: I-V measurements of a GA sensor under an increasing compressive strain demonstrating a linear relationship between current (I) and voltage (V). The slope of the I–V curves increases with increasing applied pressure.



**Figure S7**: Resistivity of graphene aerogels as a function of (a) sample annealing temperature and (b) temperature. The temperature dependent resistivity was measured using a graphene aerogel sample annealed at 1300 °C. Resistivity of the samples was determined using the van der Pauw method in a 4-point probe geometry.



**Figure S8**: (a) Photographs showing reversible mechanical and electrical nature of GA under the tensile strain (scale bar: 3 mm) and (b) the corresponding current-strain response of the GA sensor.



Figure S9: Schematics of the experimental configuration for measuring the fast dynamic voltage response of the sensors



**Figure S10**: Dynamic response of a GA sensor at (a) 15 Hz (b) 55 Hz and (c) 150 Hz at repetitive 10% compressive strain pressure conditions using an external electric motor.



**Figure S11**: Experimental set-up to excite the sensor at different frequencies of 15-150 Hz using an external electric motor showing (a) side view and (b) front view. The frequency is changed by varying the applied voltage across the motor.



**Figure S12**: Optical and SEM images of a graphene aerogel sensing element (a,c) before and (b,d) after 5000 loading-unloading cycles.

Materials	Response	Gauge Factor (GF)/ Sensitivity (S, kPa <sup>-1</sup> ) (Strain Range)	Sensing Type	Response Time	Ref
3D Graphene Aerogels	bilinear	$\begin{array}{l} GF_1 = 11.6 \; (0-8 \; kPa) \\ GF_2 = 0.38 \; (8 \; kPa-1.18 \; MPa) \\ S_1 = 53.5 \; MPa^{-1} \; (0-101 \; kPa) \\ S_2 = 12.2 \; MPa^{-1} \; (101 \; kPa-1.18 \; MPa) \\ MPa) \end{array}$	compression	0.52 ms (rise time) 0.61 ms (fall time)	This work
		$\begin{array}{l} GF_3 = 0.6 \; (0\mbox{-}0.3 \; MPa) \\ GF_4 = 3.4 \; (0.3\mbox{-}0.55 \; MPa) \\ S_3 = 0.39 \; MPa^{-1} \; (0\mbox{-}0.3 \; MPa) \\ S_4 = 1.85 \; MPa^{-1} \; (0.3\mbox{-}0.55 \; MPa) \end{array}$	tension		
Graphene foam	linear	GF = 1.3	compression	-	1
Porous GO/rGO Hybrid film	nonlinear	$S = 0.032 \text{ kPa}^{-1}(0-1 \text{ kPa})$	compression	-	2
Graphene- Based Cellular Elastomers	linear	$S_1 = 10 \text{ kPa}^{-1} (0\text{-}200 \text{ Pa})$ $S_2 = 0.23 \text{ kPa}^{-1} (0\text{-}2.7 \text{ kPa})$	compression	-	3
Graphene- amorphous carbon hierarchical foam	nonlinear	S = 11.47–5.19 kPa <sup>-1</sup> (0-18 Pa)	compression	-	4
Biomimetic Architectured Graphene aerogels	nonlinear	S = 4 (50% strain)	compression	-	5
Graphene foam	bilinear	$S = 22.8 \pm 1.3 \text{ kPa}^{-1}$ (0-10 Pa)	compression	-	6
Graphene aerogels	nonlinear	94% resistance change upon 60% compression	compression	250 ms (rise time) 150 ms (recovery time)	7
Sparkling graphene block	nonlinear	$S_1 = 229.8 \text{ kPa}^{-1} (0-0.1 \text{ kPa})$ $S_2 = 26.7 \text{ kPa}^{-1} (0.4-1 \text{ kPa})$	compression	1,085 mm/s	8
Graphene sponge	bilinear	$S_1 = 1.04 \text{ kPa}^{-1} (13-260 \text{ Pa})$ $S_2 = 0.12 \text{ kPa}^{-1} (0.26-20 \text{ kPa})$	compression	34 ms (response time) 5 ms (recovery time)	9
Graphene foam-like structure	bilinear	$\begin{split} S_1 &= 0.96 \text{ kPa}^{-1} \ (<\!50 \text{ kPa}) \\ S_2 &= 0.005 \text{ kPa}^{-1} \ (>\!50 \text{ kPa}) \end{split}$	compression	0.4–212 ms	10
Sponges coated with multiwalled carbon nanotubes and graphene	nonlinear	GF = 0.96–1.75	compression	< 750 ms	11
Graphene/silve r nanowires sponge	linear	$ \begin{array}{l} S_1 = 0.29 \ \text{kPa-1} \ (0\text{-}2.5 \ \text{kPa}) \\ S_2 = 0.02 \ \text{kPa-1} \ (3\text{-}10 \ \text{kPa}) \\ GF = 1.5 \ (0\%\text{-}60\% \ \text{strain}) \end{array} $	compression	54 ms	12
Graphene/poly urethane sponge	nonlinear	$S = 0.75 - 3.08 \text{ kPa}^{-1}$	compression	14 ms	13

**Table S4**: Comparison of our GA sensor with other reported strain-gauge sensors based on 3D graphene and other materials.

Graphene– Polyurethane	bilinear	$S_1 = 0.26 \text{ kPa}^{-1} (0-2 \text{ kPa})$ $S_2 = 0.03 \text{ kPa}^{-1} (2-10 \text{ kPa})$	compression		14
Sponge					
3D periodic	-	GF ~ 1 (50% strain)	compression	-	15
graphene					
aerogel					
microlattices					
3D-printed	linear	GF = 1.34 (up to 20% strain)	compression		16
graphene					
aerogel					
Graphene	bilinear	$S = 0.09 \text{ kPa}^{-1}$ (up to 2000 kPa)	compression	80–100 ms	17
foam/PDMS		$GF_1 = 2.6 \ (0-18\%)$			
Porous		$GF_2 = 8.5 (22 - 40\%)$			
Network					
Structure					
Conventional		Constantan wire: GF = 2.06			18,19
metal alloys		Constantan foil: $GF = 2.04-2.16$			
		NiCr. Foil: GF = 1.98-2.01			

#### References

- J. Kuang, L. Liu, Y. Gao, D. Zhou, Z. Chen, B. Han and Z. Zhang, *Nanoscale*, 2013, 5, 12171.
- 2 S. Liu, X. Wu, D. Zhang, C. Guo, P. Wang, W. Hu, X. Li, X. Zhou, H. Xu, C. Luo, J. Zhang and J. Chu, *ACS Appl. Mater. Interfaces*, 2017, **9**, 24148–24154.
- 3 L. Qiu, M. Bulut Coskun, Y. Tang, J. Z. Liu, T. Alan, J. Ding, V.-T. Truong and D. Li, *Adv. Mater.*, 2016, **28**, 194–200.
- 4 Y. Ma, M. Yu, J. Liu, X. Li and S. Li, ACS Appl. Mater. Interfaces, 2017, 9, 27127–27134.
- 5 M. Yang, N. Zhao, Y. Cui, W. Gao, Q. Zhao, C. Gao, H. Bai and T. Xie, *ACS Nano*, 2017, **11**, 6817–6824.
- 6 X. Zang, X. Wang, Z. Yang, X. Wang, R. Li, J. Chen, J. Ji and M. Xue, *Nanoscale*, 2017, **9**, 19346–19352.
- 7 K. Hu, T. Szkopek and M. Cerruti, J. Mater. Chem. A, 2017, 5, 23123–23130.
- 8 L. Lv, P. Zhang, T. Xu and L. Qu, ACS Appl. Mater. Interfaces, 2017, 9, 22885–22892.
- 9 S. Chun, A. Hong, Y. Choi, C. Ha and W. Park, *Nanoscale*, 2016, **8**, 9185–9192.
- 10 H. Tian, Y. Shu, X. F. Wang, M. A. Mohammad, Z. Bie, Q. Y. Xie, C. Li, W. T. Mi, Y. Yang and T. L. Ren, *Sci. Rep.*, 2015, **5**, 1–6.
- 11 Z. Ma, A. Wei, J. Ma, L. Shao, H. Jiang, D. Dong, Z. Ji, Q. Wang and S. Kang, *Nanoscale*, 2018, **10**, 7116–7126.
- 12 X. Dong, Y. Wei, S. Chen, Y. Lin, L. Liu and J. Li, *Compos. Sci. Technol.*, 2018, **155**, 108–116.
- 13 Y. Luo, Q. Xiao and B. Li, *RSC Adv.*, 2017, 7, 34939–34944.
- 14 H.-B. Yao, J. Ge, C.-F. Wang, X. Wang, W. Hu, Z.-J. Zheng, Y. Ni and S.-H. Yu, *Adv. Mater.*, 2013, **25**, 6692–6698.
- 15 C. Zhu, T. Y.-J. Han, E. B. Duoss, A. M. Golobic, J. D. Kuntz, C. M. Spadaccini and M. A. Worsley, *Nat. Commun.*, 2015, **6**, 6962.

- 16 Q. Zhang, F. Zhang, S. P. Medarametla, H. Li, C. Zhou and D. Lin, *Small*, 2016, **12**, 1702–1708.
- 17 Y. Pang, H. Tian, L. Tao, Y. Li, X. Wang, N. Deng, Y. Yang and T.-L. Ren, *ACS Appl. Mater. Interfaces*, 2016, **8**, 26458–26462.
- 18 K. M. B. Jansen, *Exp. Mech.*, 1997, **37**, 245–249.
- 19 M.-H. Bao, in *Handbook of Sensors and Actuators*, 2000, pp. 199–239.