Supporting Information

Highly Stretchable, Sensitive Strain Sensors with Wide Linear Sensing Region Based on Compressed Anisotropic Graphene Foam/Polymer Nanocomposites

*Zhihui Zeng, Seyed Ismail Seyed Shahabadi, Boyang Che, Youfang Zhang, Chenyang Zhao, and Xuehong Lu**

School of Materials Science and Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore
*Corresponding authors

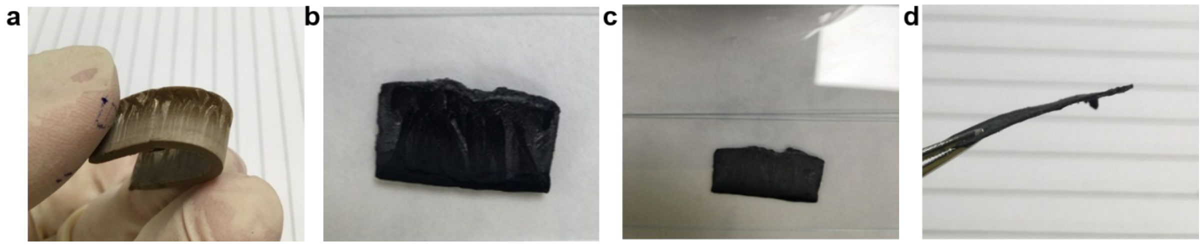

Fig. S1 Pictures showing (a) a flexible GO foam, (b) the corresponding RGO foam and (c-d) CRGO foam.

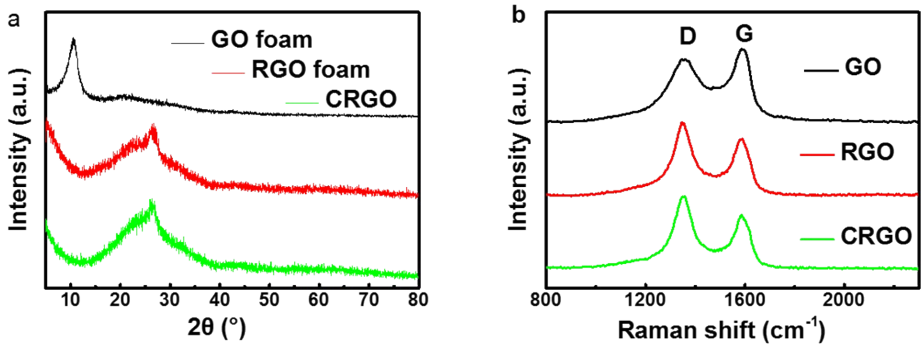

Fig. S2 (a) XRD patterns of the GO, RGO and CRGO foams, showing a strong characteristic peak at around 10.7° and no obvious graphite peak for the GO foam, and a broad peak at around 26° caused by the disorder in the graphene layers for the RGO and CRGO foams. The XRD patterns indicate the effective reduction of the GO to RGO foam. (b) Raman spectra of the GO, RGO and CRGO foams. The ratio of the I_D to I_G increases significantly from 0.86 to 1.28 when the GO is reduced to RGO, which can be attributed to the decreased average size of the sp^2 domain after thermal annealing treatment of GO. The I_D to I_G ratio remains similar when RGO is compressed to form CRGO, demonstrating insignificant structural change of the graphene layers after the compressive process. [18, 42]

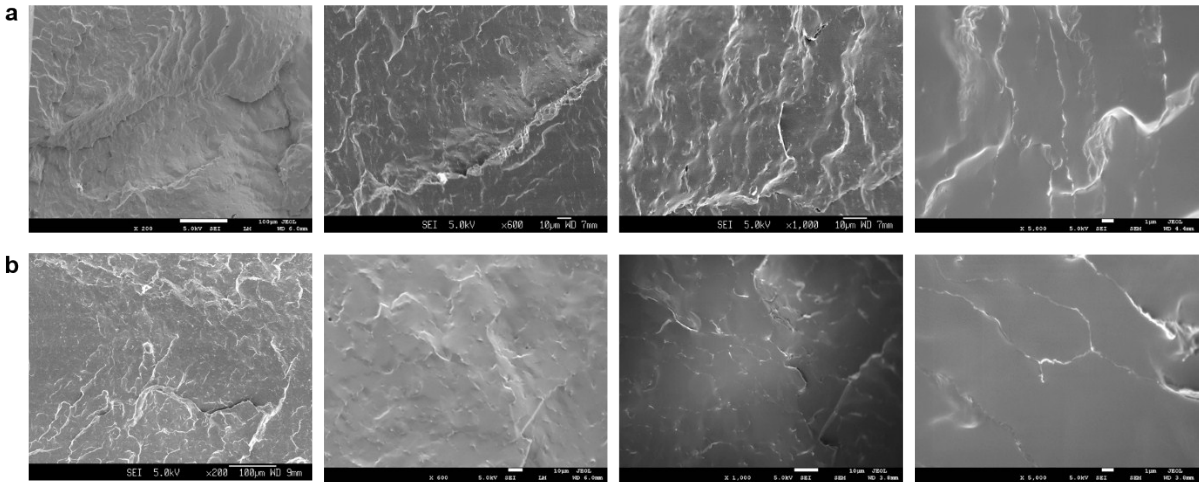

Fig. S3 Cryogenically-fractured surfaces of the CRGO/PDMS strain sensors: (a) the fractured surface parallel to the longitudinal direction with distinguishable aligned graphene layers and (b) the fractured surface parallel to the transverse direction, showing the absence of the aligned graphene layers in this plane. (Scale bars are 100, 10, 10 and 1μm from left to right columns, respectively.)

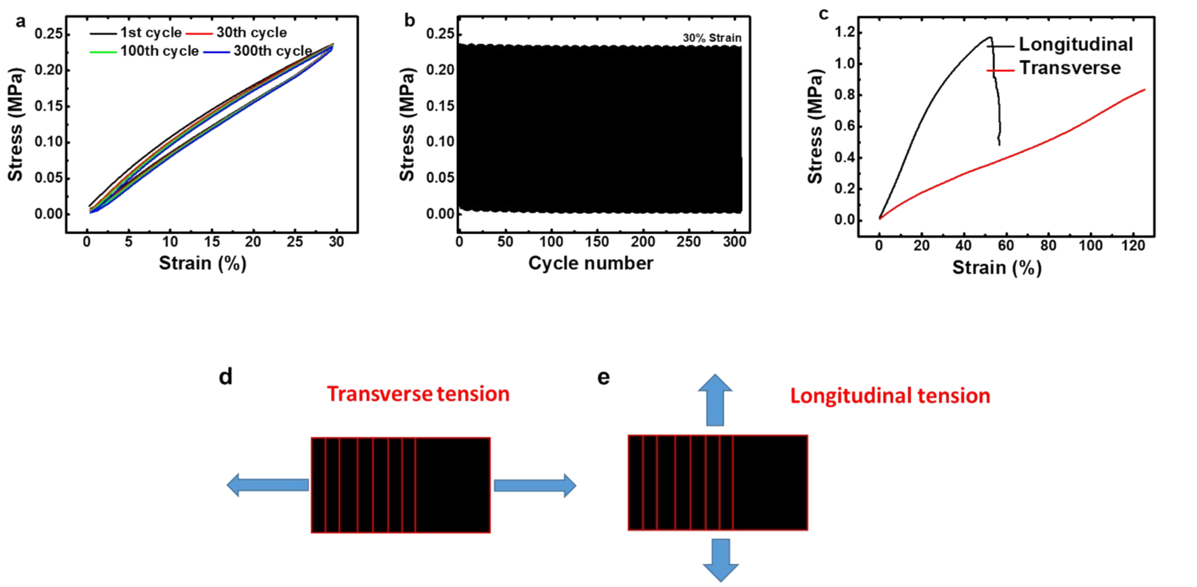

Fig. S4 (a-b) Dynamic mechanical performance of the CRGO/PDMS nanocomposite strain sensors under 30 % cyclic strain (minor hysteresis can be observed due to the mechanical properties of PDMS), (c) stress versus strain curves of the strain sensors stretched along transverse and longitudinal directions, and schematics showing the strain sensors stretched along (d) transverse and (e) longitudinal direction.

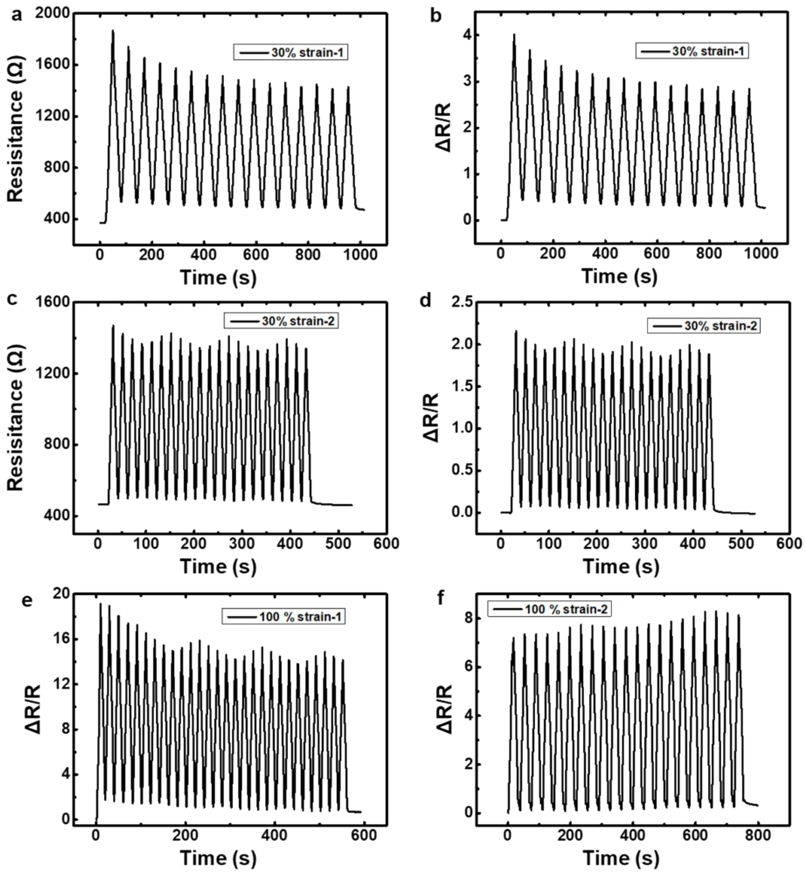

Fig. S5 (a, c) Resistance change and (b, d) relative resistance change of the CRGO/PDMS nanocomposite strain sensors at stretched strain of 30 %: (a, b) stretched from the initial state and (c, d) stretched after pre-stretching. (e, f) Relative resistance change of CRGO/PDMS nanocomposite strain sensors at stretched strain of 100 %: stretched (e) from the initial state and (f) after pre-stretching.

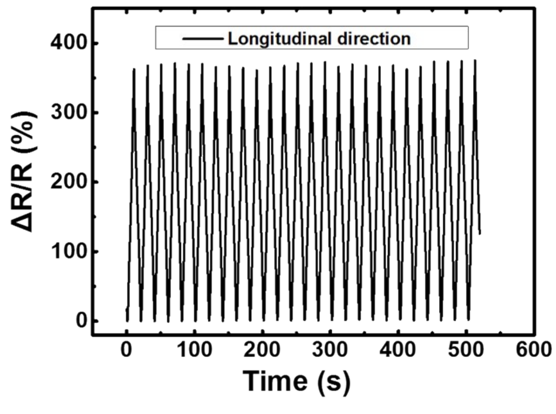

Fig. S6 Relative resistance change of the CRGO/PDMS nanocomposite strain sensors under a cyclic strain along the longitudinal direction.

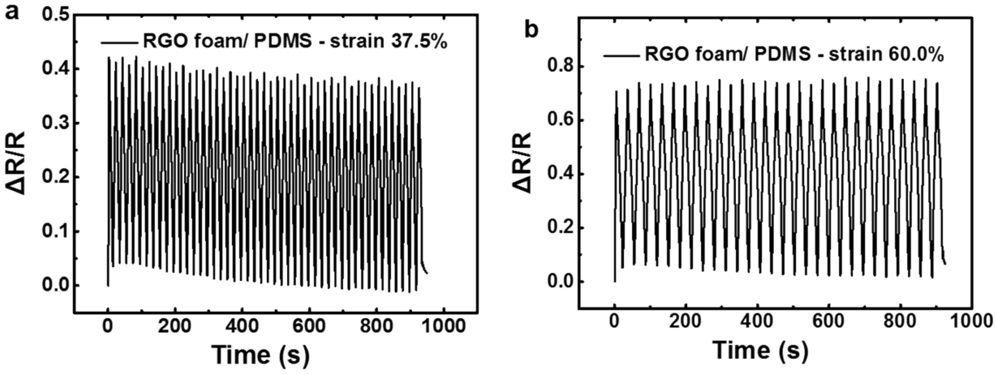

Fig. S7 Relative resistance change versus time curves of uncompressed RGO foam/PDMS nanocomposites under cyclic strains of (a) 37.5 % and (b) 60.0 %.

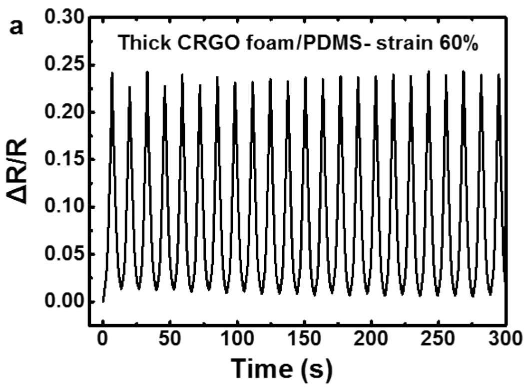

Fig. S8 Relative resistance change versus time curves of the thicker CRGO foam/PDMS nanocomposites under cyclic strain of 60 %. The foam was prepared by compressing a RGO foam with thickness of 8mm.

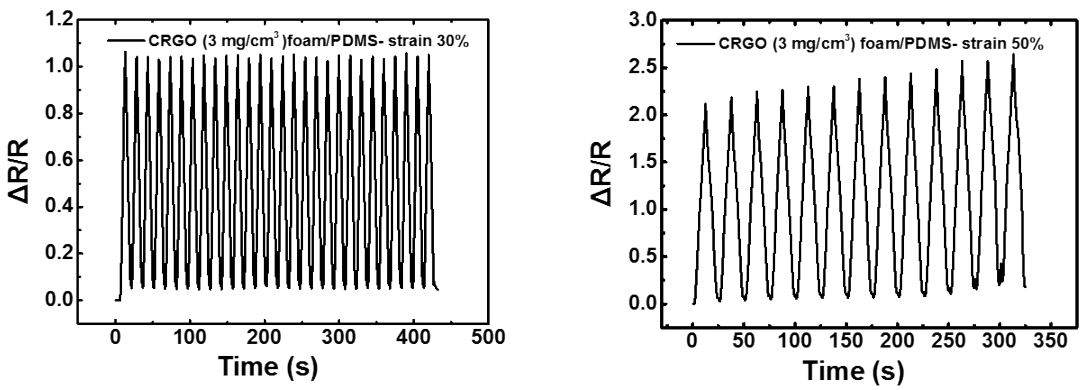

Fig. S9 Relative resistance change versus time curves of the higher-density CRGO foam /PDMS nanocomposites under cyclic strains of (a) 30 % and (b) 50 %. The foams were prepared by compressing the RGO foams with a density of 3 mg/cm^3.

Table S1. Calculated GF of a CRGO foam/PDMS nanocomposite sensor in the transverse direction at various strains.

Strain	10.0 %	27.2 %	40.8 %	54.4 %	68 %	81.6 %	95.3 %	108.9 %	122.5 %
GF	8.2	9.2	8.2	7.4	7.2	6.9	7.0	6.9	8.3

Table S2. Strain sensing performance of the graphene-based sensors

Device	Gauge factor	Cyclic strain	Linear region	Measurable strain limit	Response type	Notes
Compressed aligned porous graphene foam/PDMS	**7.2**	**100%**	**0 - 110%**	**~122%**	Linear	Transverse direction
(This work)	**12.2 27.0**	**30%**	**0 - 30%, 30% - 57%**	**~57%**	2 Linear region	Longitudinal direction
Graphene-nanocellulose /PDMS[1]	1.6 - 7.1	/		100%	Exponential	
Graphene yarn/PDMS[2]	1.4	100%	0 - 150%	150%	Linear	
Graphene fiber/PDMS[3]	3.7	50%	0 – 15%	200%	Linear & exponential	
CVD graphene foam coated with PDMS[4]	1- 2	55%		95%	Exponential	
Fish-scale-like graphene on elastic tape[5]	16.2	30%	0 - 60%	82%	Linear & exponential	
Fragmentized graphene foam/PDMS[6]	2.4-15	50%		78%	Exponential	
CVD graphene foam/PDMS[7]	2.6, 8.5	25%	0 - 18%, 22% - 40%	~48%	2 Linear & exponential	
Graphene-CNT/PDMS[8]	100	40%	10% - 40%	40%	Linear	
Graphene woven fabrics/PDMS[9]	35 -10^6	10%	/	30%	Exponential	
Graphene ripple/PDMS[10]	2	/	0 - 30%	30%	Linear	Resistance decrease
Graphene aerogel/PDMS[11]	61.3	~10%	0 - 12%	~19%	Linear & exponential	
Graphene platelets/PDMS[12]	27.7 - 164.5	10%		12.1%	Exponential	
Monolayer graphene on PDMS[13]	~151	/	3% - 4.5%	5%	/	

Suspended graphene ribbon[14]	1.9	3%		3%	Linear	
Rosette graphene/PDMS[15]	2.4	2%	0 - 2%	7.1%	Linear & exponential	Perfect reversible only under 2%
Ultrathin graphene film on PDMS[16]	228	2%	0 - 2%	~4.4%	Linear & exponential	Few-layer graphene
Percolative graphene film on PET substrate[17]	15	1.7%	0 - 1.7%	/	Linear	
Few-layer graphene on PDMS[18]	6.1	1%	0 - 1%	/	Linear	
Graphene nanoplatelet on PET[19]	8.59	0.16 %	0.1%	0.16%	Linear	
SWCNT/Graphene nanoplatelet on PET[19]	5.01	0.16 %	0.1%	0.16%	Linear	
Freestanding Graphene film[20]	/	/	/	0.85%	/	
Graphene/PVDF[21]	12.1	0.13	0 - 0.13%	/	Linear	

/: not shown, the numbers in the square brackets denote the numbers of references which are at the end of the supporting information.

1 C. Yan, J. Wang, W. Kang, M. Cui, X. Wang, C. Y. Foo, K. J. Chee, P. S. Lee, *Adv. Mater.* 2014, 26, 2022.

2 J. J. Park, W. J. Hyun, S. C. Mun, Y. T. Park, O. O. Park, *ACS Appl. Mater. Interf.* 2015, 7, 6317.

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

4 Z. Chen, W. Ren, L. Gao, B. Liu, S. Pei, H.-M. Cheng, *Nat. Mater.* 2011, 10, 424.

5 Q. Liu, J. Chen, Y. Li, G. Shi, *ACS Nano* 2016, 10, 7901.

6 Y. R. Jeong, H. Park, S. W. Jin, S. Y. Hong, S.-S. Lee, J. S. Ha, *Adv. Funct. Materi.* 2015,

25, 4228.

7 Y. Pang, H. Tian, L. Tao, Y. Li, X. Wang, N. Deng, Y. Yang, T. L. Ren, *ACS Appl. Mater. Interf.* 2016, 8, 26458.

8 C. Lee, L. Jug, E. Meng, *Appl. Phys. Lett.* 2013, 102, 183511.

9 Y. Wang, L. Wang, T. Yang, X. Li, X. Zang, M. Zhu, K. Wang, D. Wu, H. Zhu, *Adv. Funct. Mater.* 2014, 24, 4666; X. Li, R. Zhang, W. Yu, K. Wang, J. Wei, D. Wu, A. Cao, Z. Li, Y. Cheng, Q. Zheng, R. S. Ruoff, H. Zhu, *Sci. Rep.* 2012, 2, 870.

10 Y. Wang, R. Yang, Z. Shi, L. Zhang, D. Shi, E. Wang, G. Zhang, *ACS Nano* 2011, 5, 3645.

11 S. Wu, R. B. Ladani, J. Zhang, K. Ghorbani, X. Zhang, A. P. Mouritz, A. J. Kinloch, C. H. Wang, *ACS Appl. Mater. Interf.* 2016, 8, 24853.

12 G. Shi, Z. Zhao, J.-H. Pai, I. Lee, L. Zhang, C. Stevenson, K. Ishara, R. Zhang, H. Zhu, J. Ma, *Adv. Funct. Mater.* 2016, 26, 7614.

13 X.-W. Fu, Z.-M. Liao, J.-X. Zhou, Y.-B. Zhou, H.-C. Wu, R. Zhang, G. Jing, J. Xu, X. Wu, W. Guo, D. Yu, *Appl. Phys. Lett.* 2011, 99, 213107.

14 M. Huang, T. A. Pascal, H. Kim, W. A. Goddard, J. R. Greer, *Nano Lett.* 2011, 11, 1241.

15 S.-H. Bae, Y. Lee, B. K. Sharma, H.-J. Lee, J.-H. Kim, J.-H. Ahn, *Carbon* 2013, 51, 236.

16 X. Li, T. Yang, Y. Yang, J. Zhu, L. Li, F. E. Alam, X. Li, K. Wang, H. Cheng, C.-T. Lin, Y. Fang, H. Zhu, *Adv. Funct. Mater.* 2016, 26, 1322.

17 M. Hempel, D. Nezich, J. Kong, M. Hofmann, *Nano Lett.* 2012, 12, 5714.

18 Y. Lee, S. Bae, H. Jang, S. Jang, S. E. Zhu, S. H. Sim, Y. I. Song, B. H. Hong, J. H. Ahn, *Nano Lett.* 2010, 10, 490.

19 S. Luo, T. Liu, *Adv. Mater.* 2013, 25, 5650.

20 X. Xie, H. Bai, G. Shi, L. Qu, *J. Mater. Chem.* 2011, 21, 2057.

21 V. Eswaraiah, K. Balasubramaniam, S. Ramaprabhu, *Nanoscale* 2012, 4, 1258.