

## Supporting information

# Highly stretchable conductive MWCNT-PDMS composite with self-enhanced conductivity

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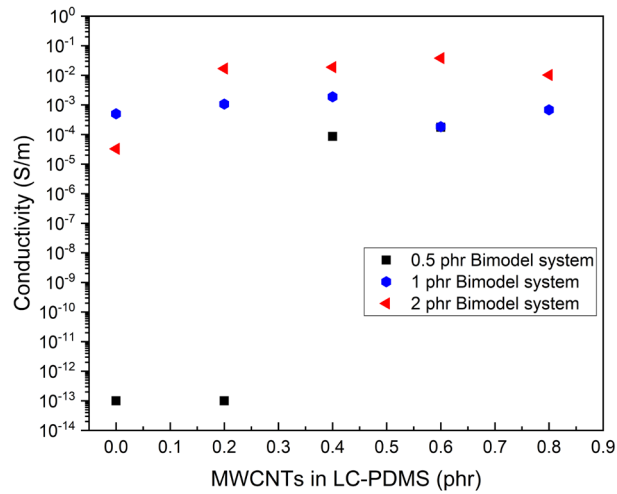
Fig. S8 dielectric spectroscopy of MWCNTs/PDMS composites.

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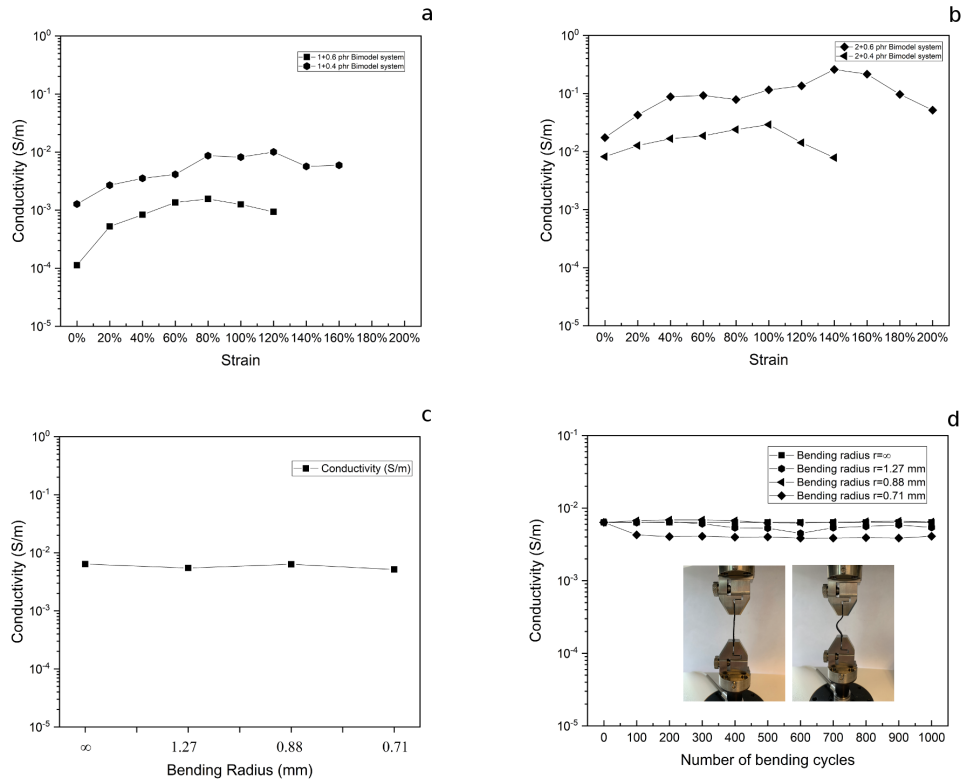
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**Table S1:** The dependence of conductivity on concentration of MWCNTs in SC-PDMS and LC-PDMS.

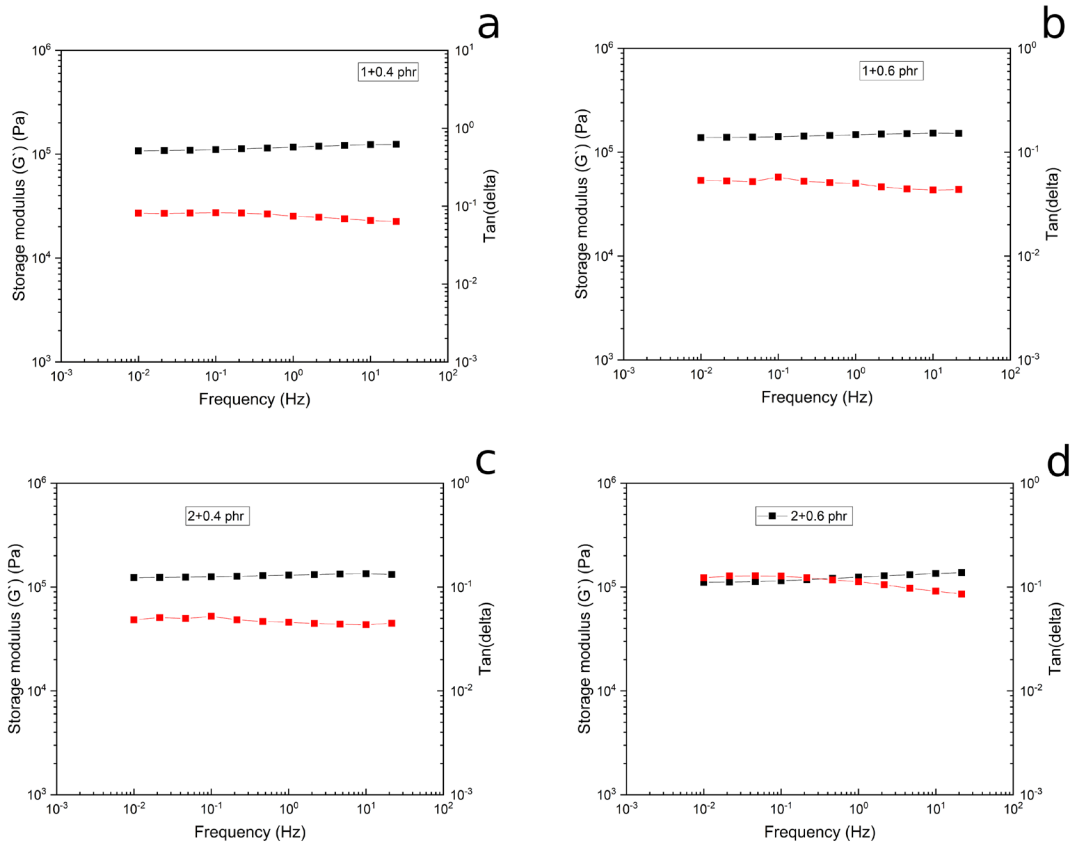
Conductivity (S/m)	MWCNTs from LC-domain (phr, overall)				
MWCNTs from SC-domain (phr, overall)	0	0.2	0.4	0.6	0.8
0.5	0	0	$8.65 \times 10^{-5}$	$1.75 \times 10^{-4}$	---
1	$5.03 \times 10^{-4}$	$1.07 \times 10^{-3}$	$1.88 \times 10^{-3}$	$1.83 \times 10^{-4}$	$6.83 \times 10^{-4}$
2	$3.28 \times 10^{-5}$	$1.69 \times 10^{-2}$	$1.89 \times 10^{-2}$	$3.81 \times 10^{-2}$	$1.03 \times 10^{-2}$



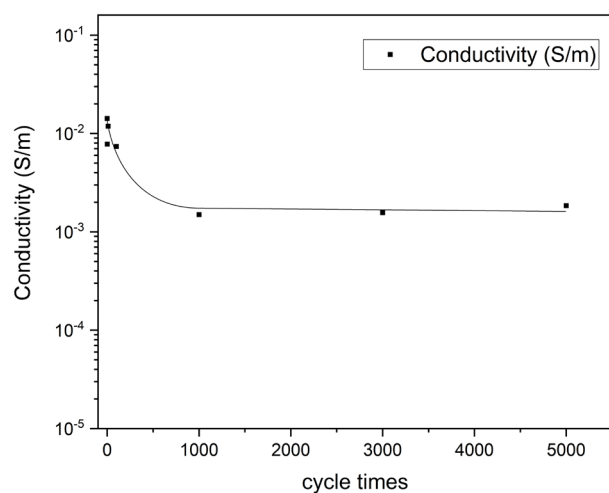
**Fig. S1** the dependence of conductivity on concentration of MWCNTs in SC-PDMS and LC-PDMS.



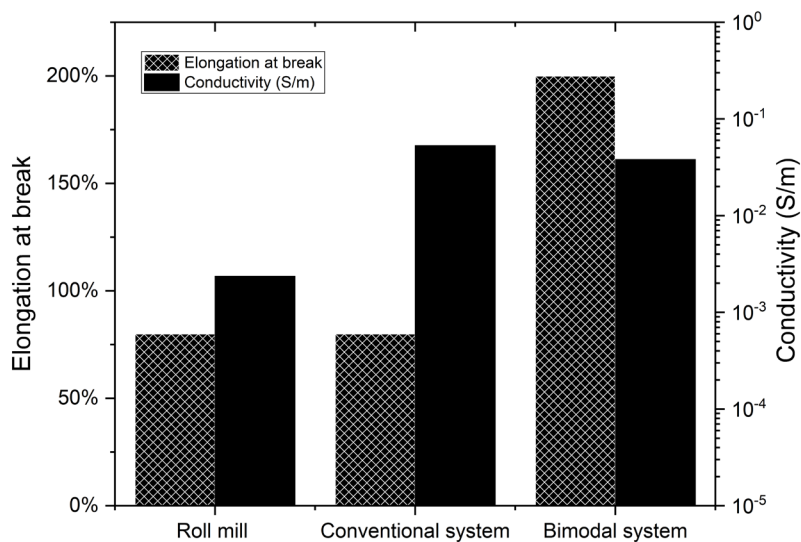
**Fig. S2** Electrical properties of MWCNTs/PDMS composites and reference samples: a) Conductivity of MWCNTs/PDMS composites with 1+0.4 phr and 1+0.6 phr MWCNTs under different strains. b) Conductivity of MWCNTs/PDMS composites with 2+0.4 phr and 2+0.6 phr MWCNTs under different strains. c) Conductivity of MWCNTs/PDMS composites with 2+0.6 phr MWCNTs at different bending radiuses. d) Conductivity of MWCNTs/PDMS composites with 2+0.6 phr MWCNTs at different bending radiuses and bending cycles.



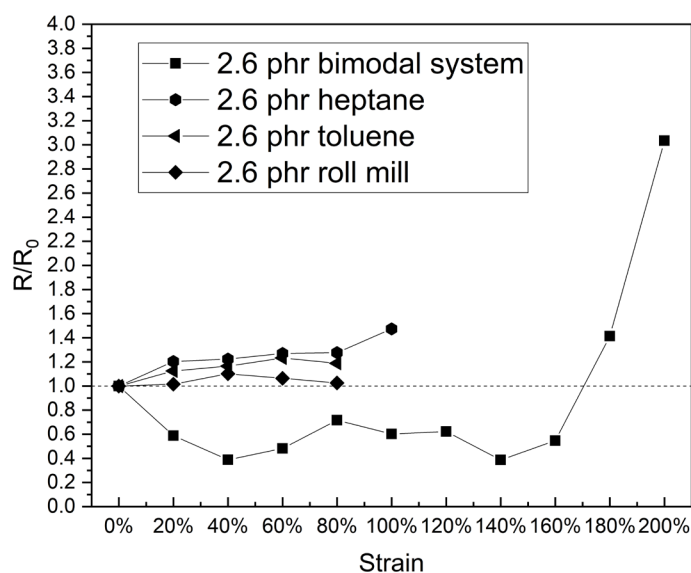
**Fig. S3** rheological properties of MWCNTs/PDMS composites: a) rheological properties of MWCNTs/PDMS composites with 1+0.4 phr MWCNTs b) rheological properties of MWCNTs/PDMS composites with 1+0.6 phr MWCNTs. c) rheological properties of MWCNTs/PDMS composites with 2+0.4 phr MWCNTs. d) rheological properties of MWCNTs/PDMS composites with 2+0.6 phr MWCNTs.



**Fig. S4** electrical conductivity of MWCNTs/PDMS composites with 2+0.6 phr MWCNTs as a function of stretching–releasing cycles with 30% strain



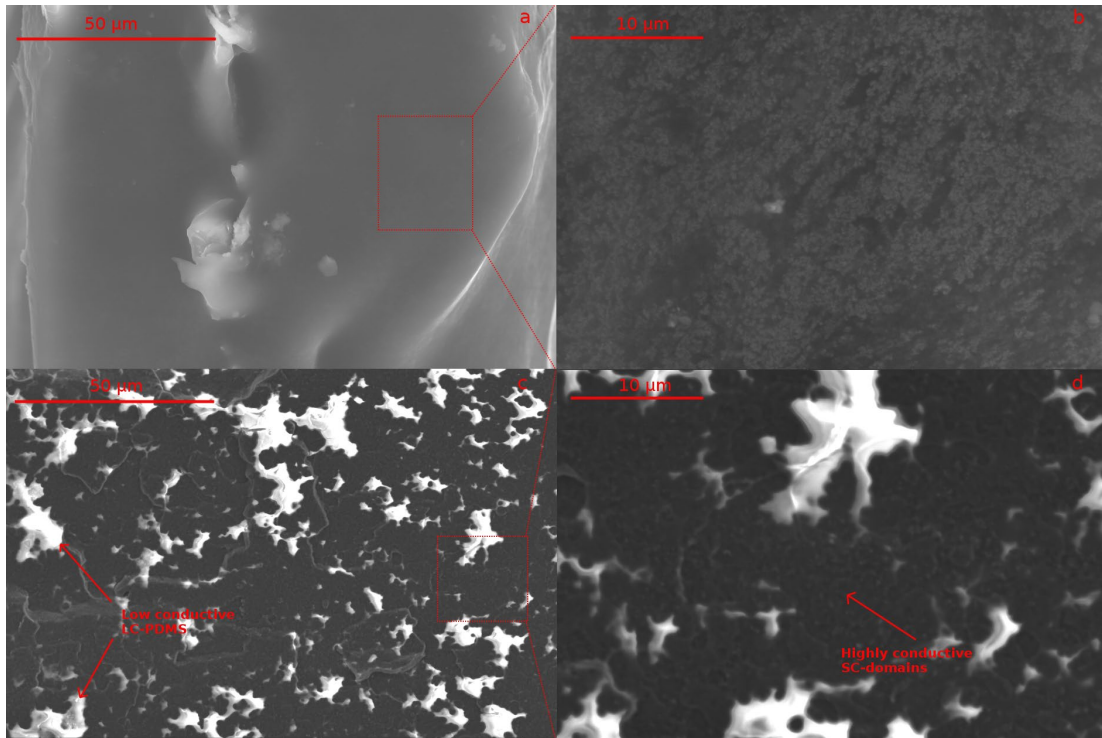
**Fig. S5** Stretchability and conductivity of samples prepared by different methods (2.6 phr MWCNTs).



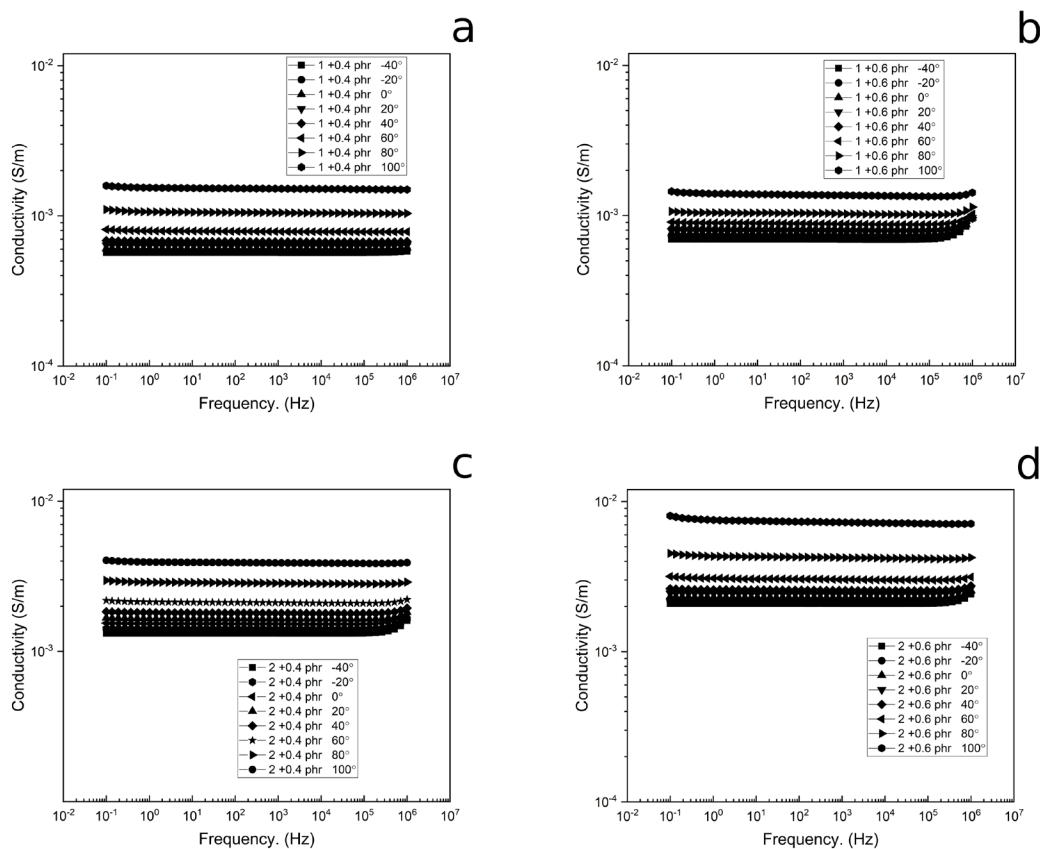
**Fig. S6** Behaviour of normalized resistances of MWCNTs/PDMS composites and reference samples under different strains (All samples are from a formulation point of view identical but the preparation schemes are varied).

**Table. S2** comparison of our results with other published results.

Fillers/polymer	Filler content	Conductivity (S/m)	Reference
MWCNT/PDMS	3 wt%	$\sim 1 \times 10^{-3}$	Hassouneh S S, et al.[1]
MWCNT/PDMS	1 wt%	$\sim 1 \times 10^{-6}$	Goswami K, et al.[2]
SWCNT/epoxy	5 wt%	$\sim 1 \times 10^{-3}$	Kim B, et al.[3]
SWCNT/polystyrene	8.5 wt%	$1.3 \times 10^{-5}$	Barraza H J, et al.[4]
MWCNT/PDMS	2.3 wt% (2.6 phr)	$3.8 \times 10^{-2}$	Shao J, et al. (This work)



**Fig. S7:** SEM micrographs of uniformly dispersed MWCNT in PDMS at 2.6 phr (a, b), and the corresponding bimodal MWCNT/PDMS system with 2+0.6 phr MWCNT (c, d). The anisotropic distribution from the bimodal mixing process is clear from the mixture of highly conductive and isolating regions in the sample.



**Fig. S8** dielectric spectroscopy of MWCNTs/PDMS composites: a) dielectric properties of MWCNTs/PDMS composites with 1+0.4 phr MWCNTs b) dielectric properties of MWCNTs/PDMS composites with 1+0.6 phr MWCNTs. c) dielectric properties of MWCNTs/PDMS composites with 2+0.4 phr MWCNTs. d) dielectric properties of MWCNTs/PDMS composites with 2+0.6 phr MWCNTs.

#### References:

- [1] S.S. Hassouneh, L. Yu, A.L. Skov, A.E. Daugaard, Soft and flexible conductive PDMS/MWCNT composites, *J. Appl. Polym. Sci.* 134 (2017) 1–9. <https://doi.org/10.1002/app.44767>.
- [2] K. Goswami, A.E. Daugaard, A.L. Skov, Dielectric properties of ultraviolet cured poly(dimethyl siloxane) sub-percolative composites containing percolative amounts of multi-walled carbon nanotubes, *RSC Adv.* 5 (2015) 12792–12799. <https://doi.org/10.1039/c4ra14637a>.
- [3] B. Kim, J. Lee, I. Yu, Electrical properties of single-wall carbon nanotube and epoxy composites, *J. Appl. Phys.* 94 (2003) 6724–6728.
- [4] H.J. Barraza, F. Pompeo, E.A. O’Rea, D.E. Resasco, SWNT-filled thermoplastic and elastomeric composites prepared by miniemulsion polymerization, *Nano Lett.* 2 (2002) 797–802.