

1 *Supporting information for*

2 **Highly conductive and flexible polymer composites**
3 **with improved mechanical and electromagnetic**
4 **interference shielding performances**

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1 **MWCNTs-carbon aerogel (MCA)/PDMS Composite Films Fabrication and**

2 **Characterization:** Without dipping the quartz fiber cloths (QFCs) into MWCNTs/resorcinol (R)/formaldehyde

3 (F) aqueous solution, the organic monomers went through a simple condensation polymerization process upon

4 nanotubes and cross-linked in gels. Thus the nanotubes network can be fixed and strengthened. After freeze-drying

5 and being pyrolyzed at 1000 °C under inert atmosphere, the gel was transformed into MCA. It is obvious that large

6 amounts of carbon nanoparticles coat on the fiber and MWCNTs (Fig. S1a). Fig. S1b shows Raman spectra for

7 pristine carbon aerogel (CA) materials coming from RF organic precursors and MWCNTs after pyrolysis. The CA

8 and MWCNTs both exhibit strong D-band and G-band peaks, which represent the disordered and ordered carbon in

9 graphitic materials, respectively.¹ In addition, the intensity ratio of D bands to G bands (I_D/I_G) for MWCNTs after

10 high-temperature treatment (HTT) is measured to be 0.68 (0.95 for pristine MWCNTs), implying an ordered

11 structure of MWCNTs. It can be seen that the pyrolysis process had little impact on the structure of nanotubes. That

12 is to say, after high-temperature treatment, the RF organic precursors transformed into graphitic materials without

13 affecting the properties of MWCNTs.

14 The aerogel/elastomer conductive composite was fabricated through vacuum impregnation of MCA with PDMS.

15 The MCA/PDMS has an electrical conductivity of 1.85 S cm⁻¹ with only 1.2 wt% MWCNTs loading, which is better

16 than QFCs-reinforced MCA/PDMS laminate (QMCA/PDMS, 1.67 S cm⁻¹, 1.6 wt% MWCNTs). But the electrical

17 conductivity retention rate of QMCA/PDMS is superior to MCA/PDMS laminate. The unique synergistic

18 MCA/QFCs structure endows QMCA/PDMS with significantly improved electrical and mechanical properties,

19 which will broaden the applications of flexible conductive materials to a great extent.

20 **QC (MWCNTs simply deposited on the QFCs without forming aerogel networks)/PDMS**

21 **Composite Films Characterization:** The conductive pathway in QC is inferior to that in QMCA preforms,

22 leading to a poor electrical conductivity of QC/PDMS, which was not remarkably improved until the MWCNTs

23 concentration reached ~ 0.5 wt% (1×10^{-3} S/cm). Fig. S3 shows the conductivity as a function of MWCNTs

24 concentration for QC/PDMS composite. Interestingly, the conductivity reaches 0.01 S/cm with 1.6 wt% MWCNTs

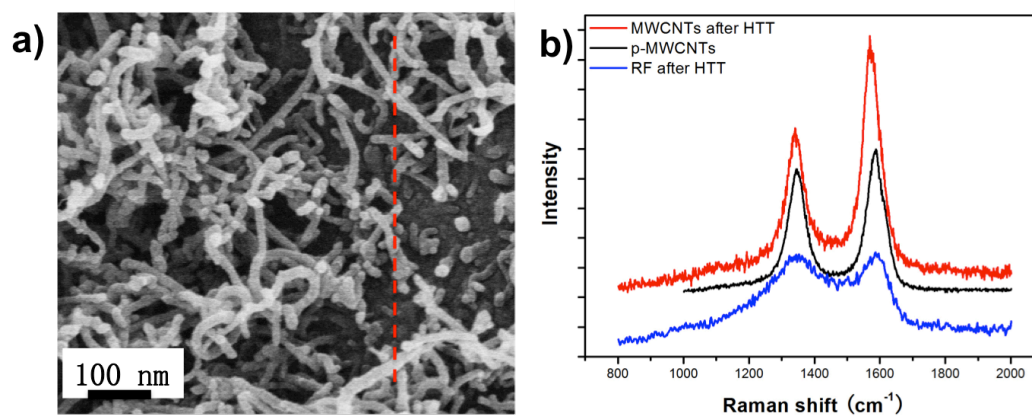
25 loading, almost two orders of magnitude smaller than QMCA/PDMS with the same MWCNTs loading (1.67 S cm⁻¹,

26 1.6 wt% MWCNTs). Moreover, to achieve the latter value, the QC/PDMS composite needs 13.5 wt% mass loading

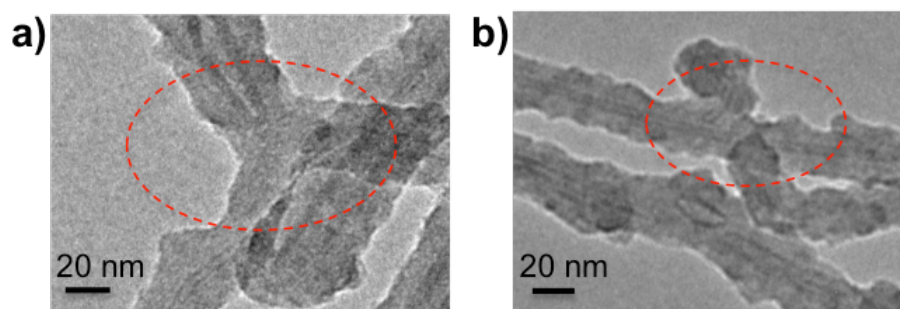
27 of nanotubes.

28 **The electromagnetic interference shielding effectiveness (EMI SE) of QMCA/PDMS and**

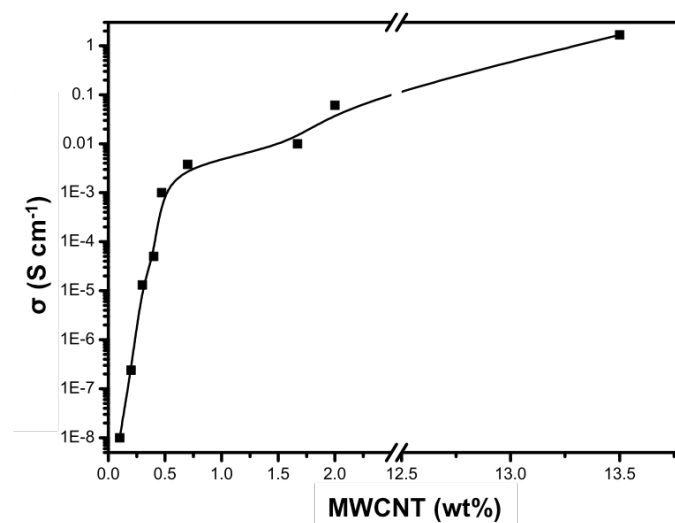
1 **QC/PDMS laminates:** The EMI SE of QMCA/PDMS and QC/PDMS laminates both increase with increased
2 nanotubes loading, as shown in Fig. S4. It is clear that the QMCA/PDMS laminates possess much better SE than
3 QC/PDMS. The SE of QC/PDMS laminate reaches the top value with 13.5 wt% MWCNTs loading, while the
4 QMCA/PDMS only needs 0.8 wt% nanotubes to achieve the same value.



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6 **Fig. S1.** a) FE-SEM image of QMCA preform, the red line represent axis direction of fiber. b) Raman spectra for MWCNTs after
7 high-temperature treatment (HTT) (red), pristine MWCNTs (p-MWCNTs, black) and RF organic aerogel after HTT (blue).



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9 **Fig. S2.** FE-TEM images of the nanotubes with CNPs coating in QMCA preform.



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11 **Fig. S3.** Conductivity of QC/PDMS composite with MWCNTs loading.

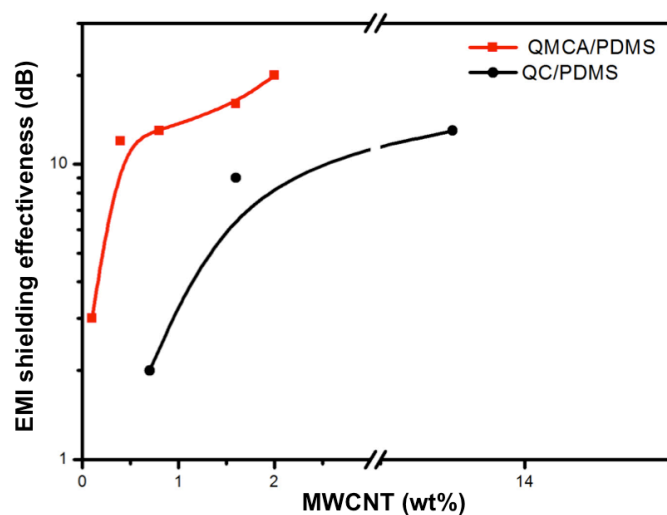


Fig. S4. The EMI SE of QMCA/PDMS and QC/PDMS composites as a function of MWCNTs loading.

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References

[1] M. A. Worsley, P. J. Pauzauskie, S. O. Kucheyev, J. M. Zaug, A. V. Hamza, J. H. Satcher Jr. and T. F. Baumann, *Acta Materialia* 2009, **57(17)**, 5131-5136.