Supporting Information

Making carbon nanotube films viscoelastic, ductile, and repairable by forming a double network with branched polyethylenimine

Xiaohua Zhang,[1]* Xin Wang,[1] Xin Zhang,[2] Jingyun Zou,[2,3]* Yongyi Zhang,[2,4] Jingna Zhao,[2,4]* Qingwen Li[2]

[1] Innovation Center for Textile Science and Technology, College of Textiles, and MOE Key Laboratory of Textile Science & Technology, Donghua University, Shanghai 201620, China.
[2] Advanced Materials Division, and Key Laboratory of Multifunctional Nanomaterials and Smart Systems, Suzhou Institute of Nano-Tech and Nano-Bionics, Chinese Academy of Sciences, Suzhou 215123, China.
[3] School of Physical Science and Technology, Suzhou University of Science and Technology, Suzhou 215009, China.
[4] Division of Nanomaterials, and Jiangxi Key Lab of Carbonene Materials, Jiangxi Institute of Nanotechnology, Nanchang, 330200, China.

*Corresponding author: Xiaohua Zhang, zhangxh@dhu.edu.cn; Jingyun Zou, zhangxh@dhu.edu.cn; Jingna Zhao, zhangxh@dhu.edu.cn

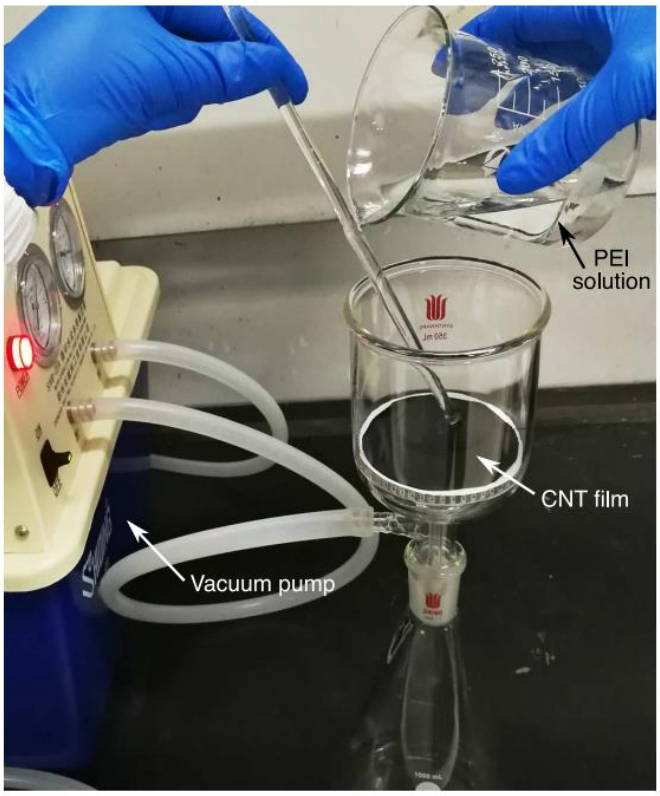

Fig. S1: Experimental set-up for the vacuum infiltration of PEI into CNT films.

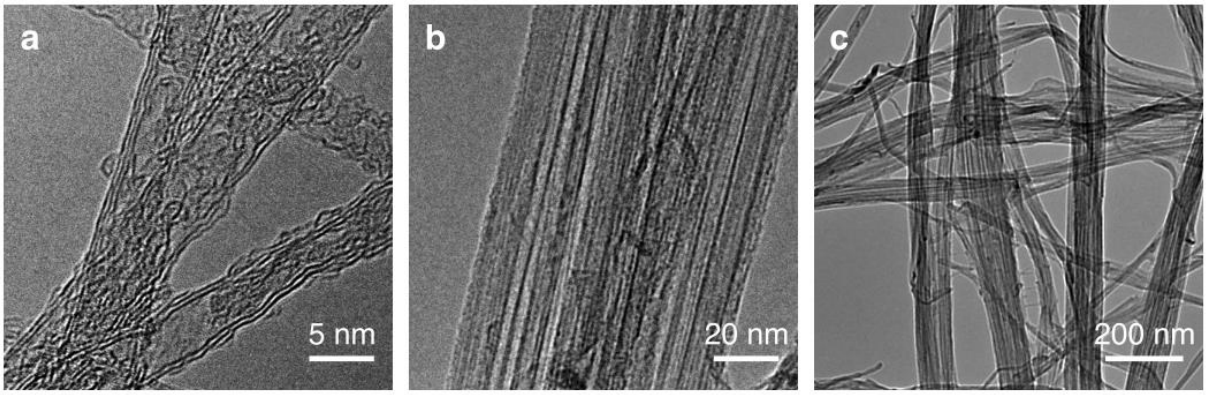

Fig. S2: TEM images of the FCCVD grown CNTs. (a) The CNTs were mainly double and triple walled. (b) The bundle size was about 80–90 nm in diameter. (c) The CNT bundles were entangled with each other.

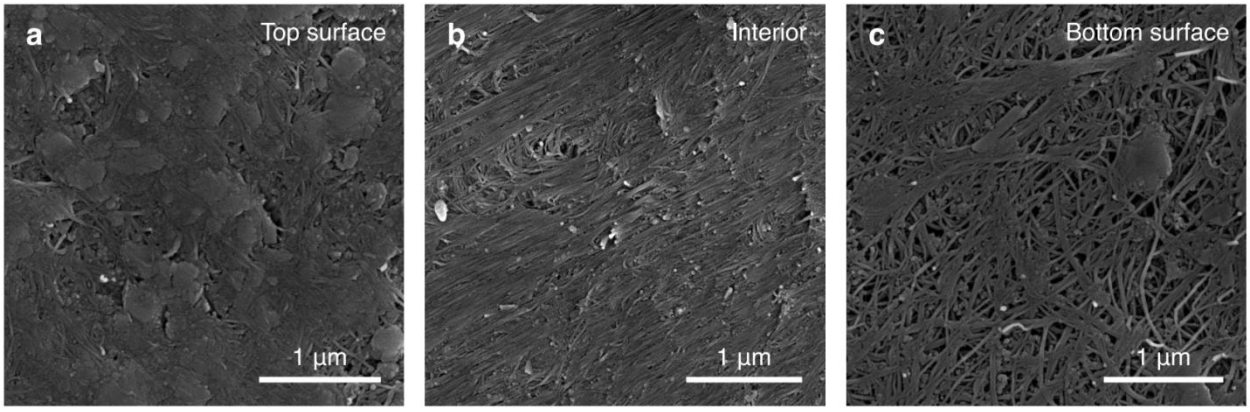

Fig. S3: SEM images showing the morphologies of the top surface (a), interior (b), and bottom surface (c) of the CNT/PEI film infiltrated with the 0.2 wt% solution. The bottom morphology indicates that PEI can well infiltrate across the CNT film. However, under such concentration, PEI was easily to be over-loaded.

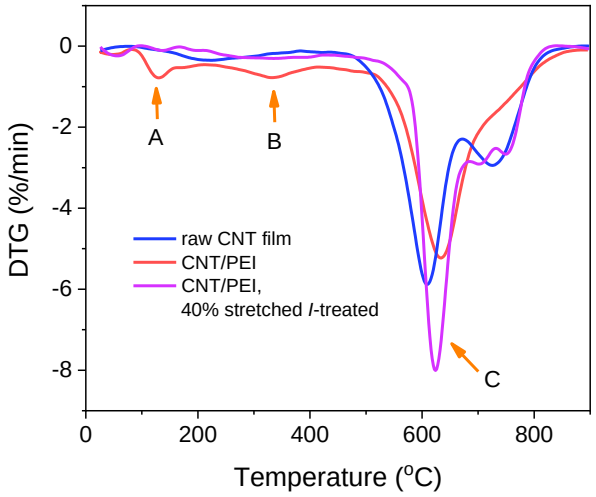

Fig. S4: DTG curves of the raw CNT film, CNT/PEI film, and the stretched and *I*-treated CNT/PEI film collected under nitrogen atmosphere. Peaks occurred around 120 °C, 330 °C, and 600 °C might be caused by the evaporation of absorbed organic byproducts as reported by our previous work, the evaporation of PEI molecules, and the decomposition of CNTs, respectively.

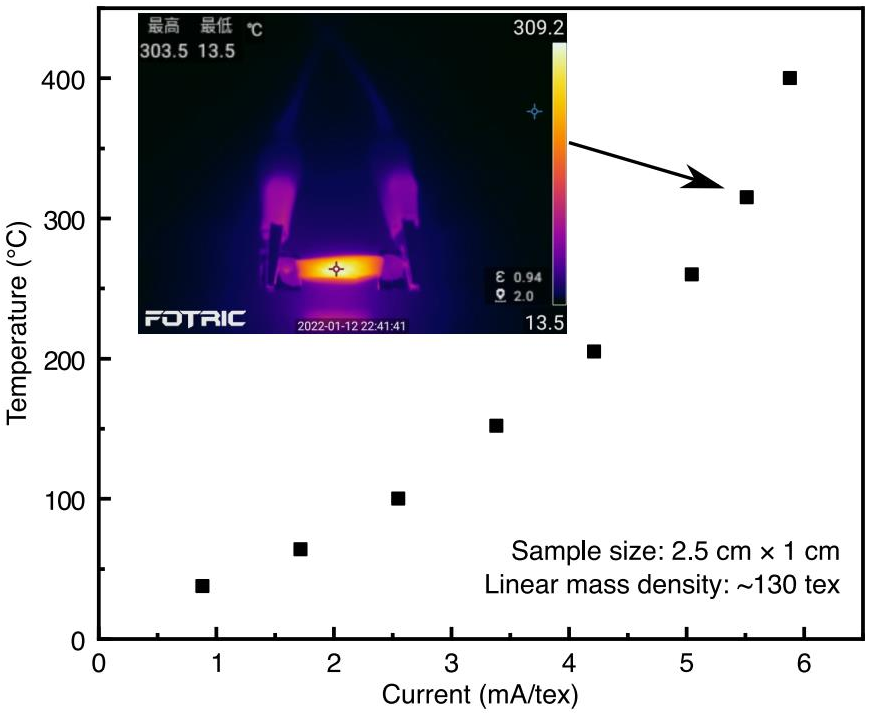

Fig. S5: Electric-heating of CNT strips. By conducting a dc current, the temperature of CNT film would increase due to the Joule effect. At a current density of about 5.5 mA tex^{-1}, the temperature could reach ~300 ◦C, high enough to remove PEI molecules inside CNT films.

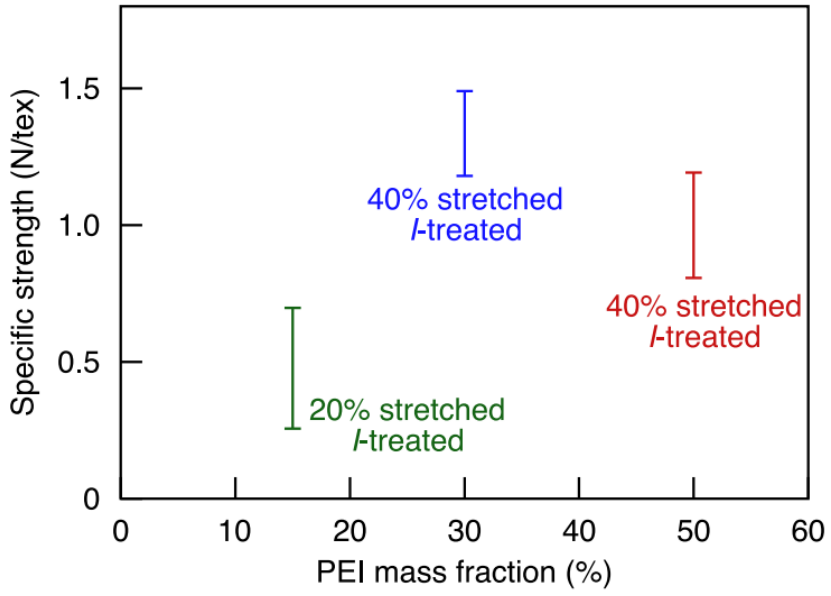

Fig. S6: Effect of PEI mass fraction on the final tensile strength of *I*-treated films. The low PEI loading (15 wt%) could not lead to high stretchability, thus the stretching degree was set to 20%. The strengths were 0.26–0.70, 1.18–1.49, and 0.81–1.19 N tex^{-1}, respectively.

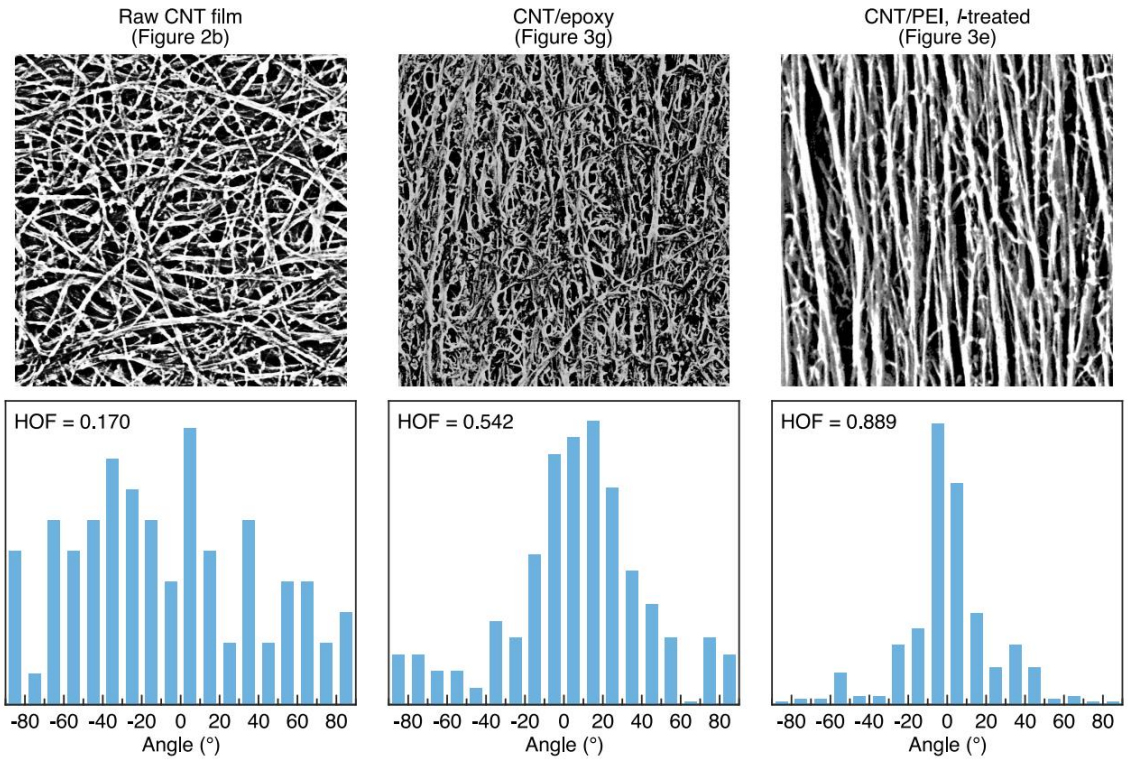

Fig. S7: Herman's orientation factors (HOF) for the raw CNT film, stretched CNT/epoxy film, and stretched CNT/PEI film, respectively. The calculation of HOF is based on the intensity profile I of orientation angle ϕ between the structural unit vector and the reference direction, according to its definition:

HOF $= 1/2\,(3\langle cos^2\,\phi\rangle - 1)$, where $\langle cos^2\,\phi\rangle = \left(\int_0^{\pi/2} I(\phi)\,cos^2\,\phi\,\sin\phi d\phi\right)\left(\int_0^{\pi/2} I(\phi)\,\sin\phi d\phi\right)$.

Here, the intensity profile was obtained by using the 'Orientation' property of 'regionprops' in Matlab, where an SEM image should be first converted to be strongly contrasted.

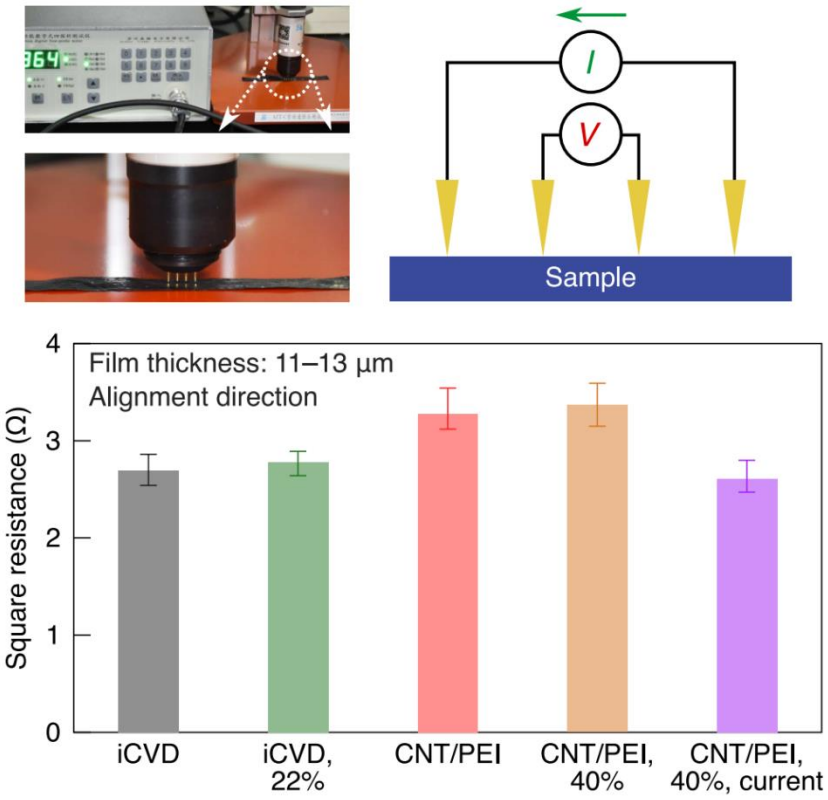

Fig. S8: Square resistance of different films, measured along the alignment direction. The infiltrated PEI could block the intertube electron hopping and thus increased the resistance. With considering the film thickness, the final conductivity of the stretched and *I*-treated film was about 3.4×10^4 S m^{-1}.

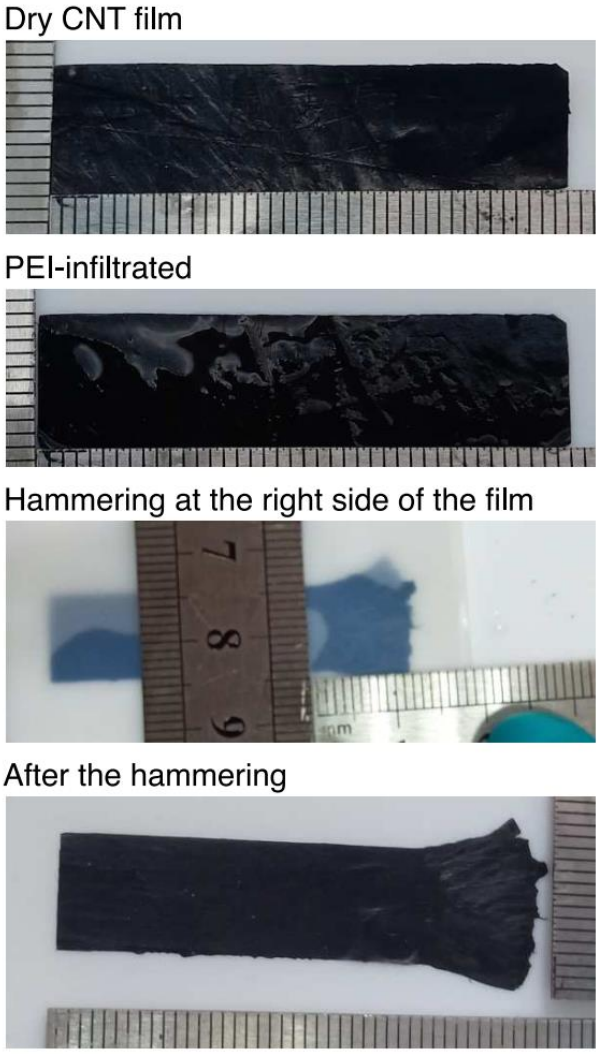

Fig. S9: Optical microscopy images showing the hammering process. It is well revealed that the CNT/PEI film exhibited superior ductility. By hammering the right side of the composite film, both the length and width dimensions could extend by more than 30%.

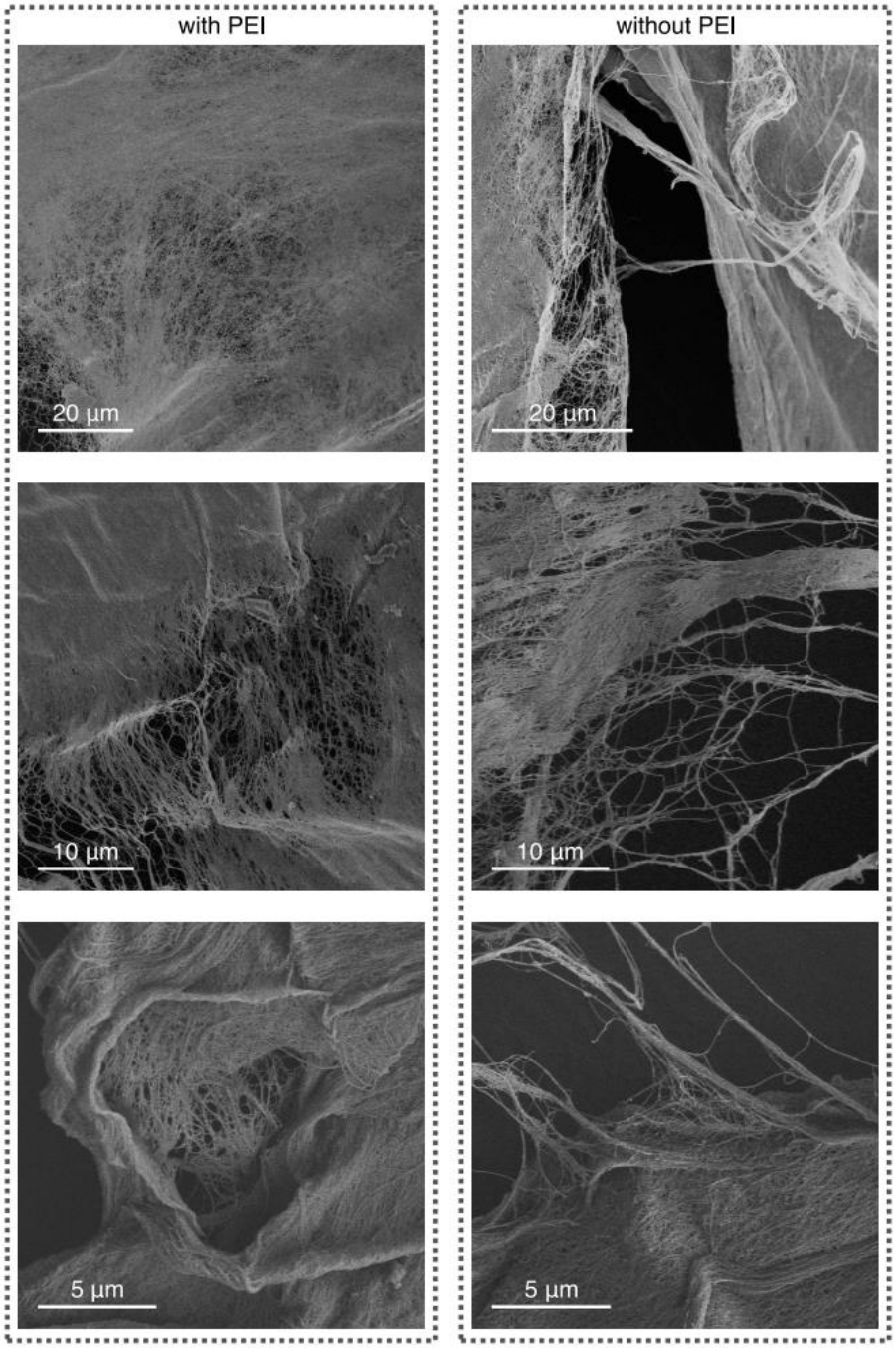

Fig. S10: Microstructures of CNT films with and without PEI after being "hammered" into ultrathin films. The presence of PEI increased the film ductility, while without PEI the film was very fragile, causing the formation of many holes.