Supporting information for

Milliseconds Tension-annealing for Enhancing Carbon Nanotube Fibers

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Keywords: carbon nanotubefibers, ultrafast Joule heating, covalent welding, high-strength, highly aligned, continuous

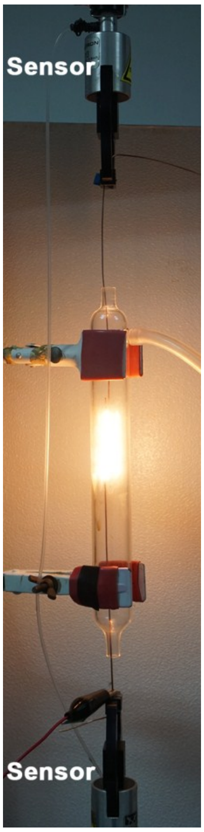

Fig.S1 (a) The in-situ measuring setup for the characterization of electromechanical response property.

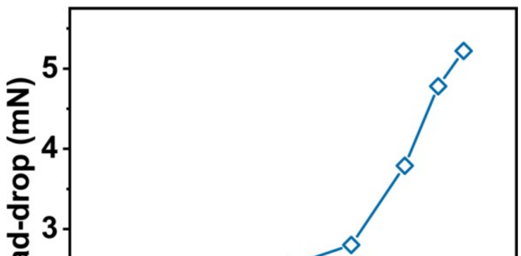

Fig.S2 The load-drops as a function of the applied tensile load during in-situ measurement of mechanical responses at the applied pulsed voltage of 70 V.

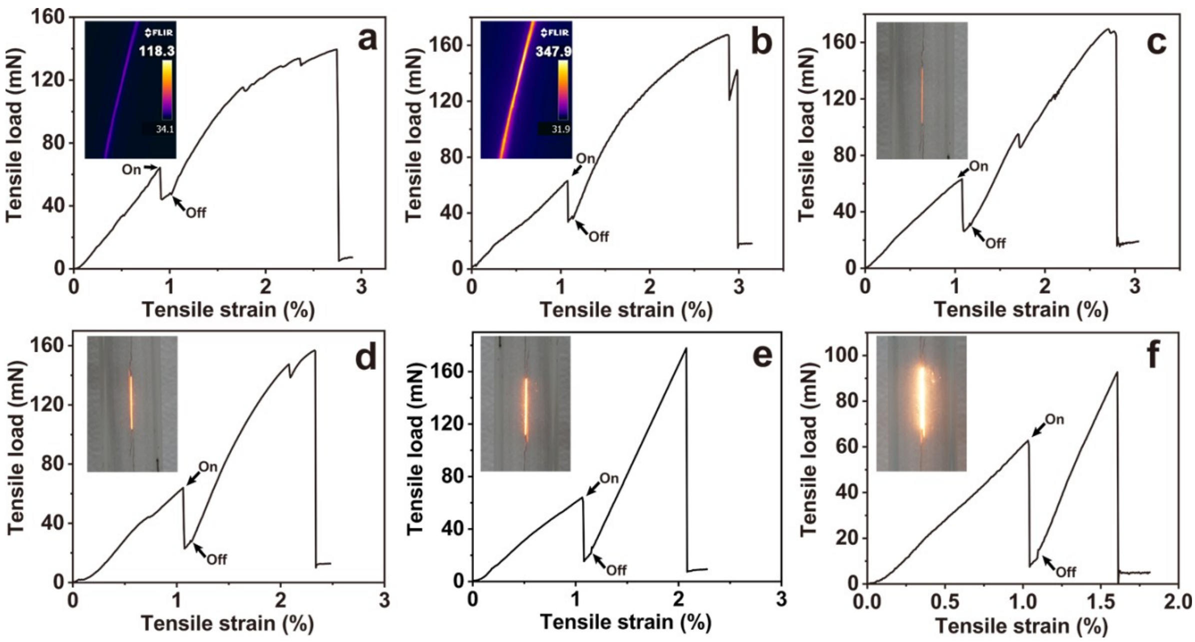

Fig.S3 (a) ~ (f) illustrate the mechanical responses of CNT fibers during tensile test when different pulsed voltages (20 V, 30 V, 40 V, 50 V, 60 V, 70 V) were applied at the tensile load of about 60 mN.

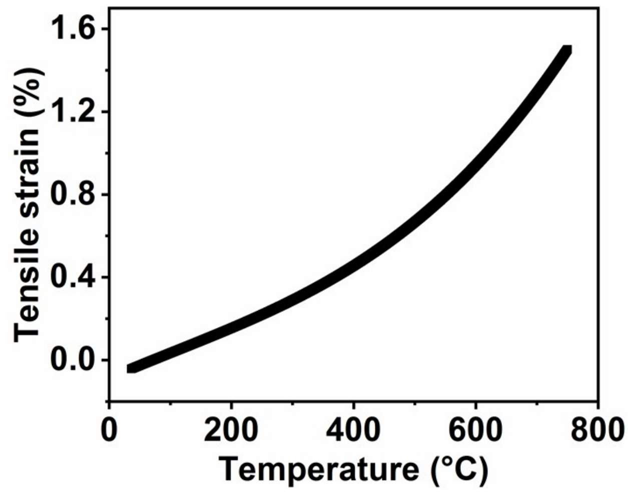

Fig.S4 Thermal expansion of pristine CNT fibers measured by thermal mechanical analyzer.

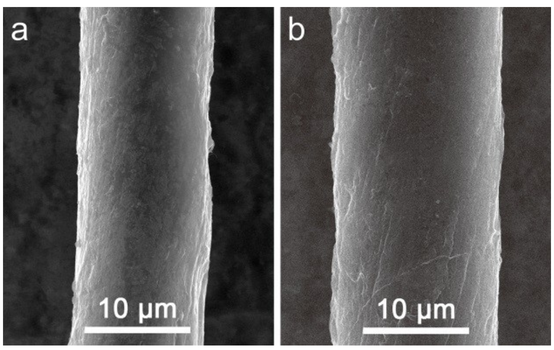

Fig.S5 (a) and (b) SEM images of pristine CNT fibers at different positions in the same batch of sample, respectively.

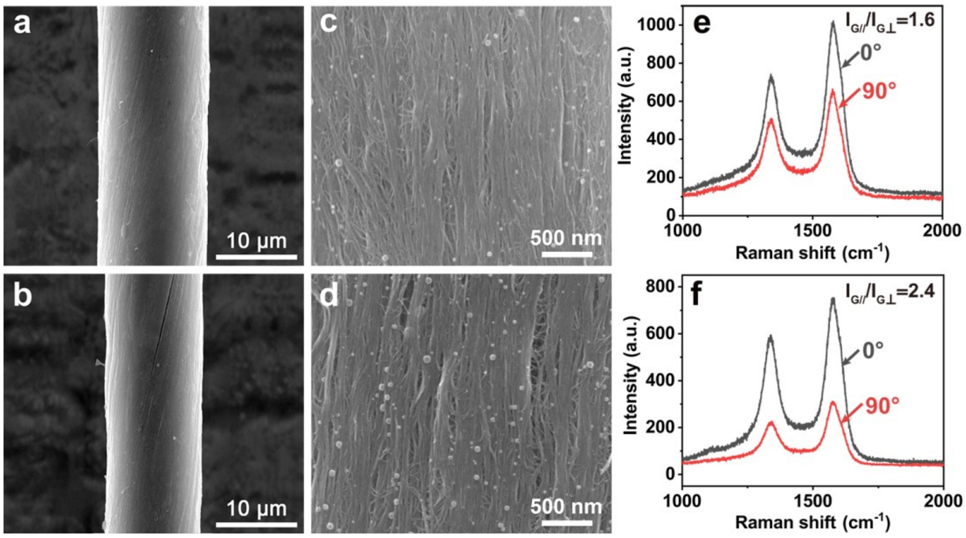

Fig.S6 (a) and (b) SEM images of pristine CNT fibers, annealed CNT fibers heated by a 50 V voltage pulse of 5 s width under the applied tensions of 0 N and 0.02 N at low magnifications, respectively. (c) and (d) SEM images of pristine CNT fibers, annealed CNT fibers heated by a 50 V voltage pulse of 5 s width under different applied tensions (0 N and 0.02 N) at high magnifications, respectively. (e) and (f) the polarized Raman spectra of D and G bands for the pristine CNT fibers, annealed CNT fibers heated by a 50 V voltage pulse of 5 s width under the applied tensions of 0 N and 0.02 N, respectively.

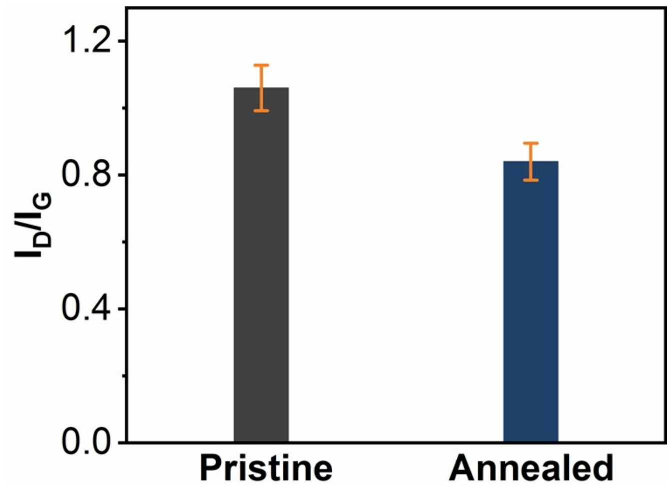

Fig.S7 I_D/I_G for pristine CNT fibers and the annealed CNT fibers.

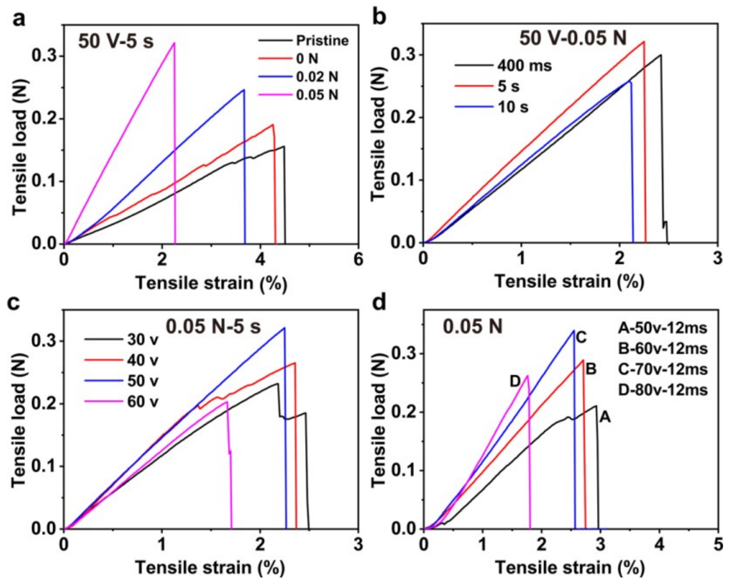

Fig.S8 (a) Load-strain curves of pristine CNT fibers and the annealed fibers at different tensile tensions (0 N, 0.02 N and 0.05 N). (b) Load-strain curves of the fibers annealed by the pulsed voltage of 50 V for different annealing times (400 ms, 5 s and 10s) under the annealing tension of 0.05 N. (c) Load-strain curves of the fibers annealed by different voltages (30 V, 40 V, 50 V and 60 V) for 5 s under the annealing tension of 0.05 N. (d) Load-strain curves of the fibers annealed by different voltages (50 V, 60 V, 70 V and 80 V) for 12 ms under the annealing tension of 0.05 N.

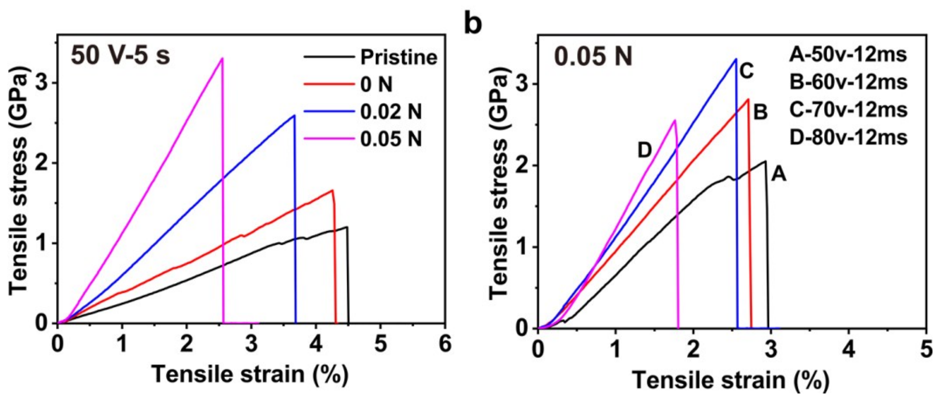

Fig.S9 (a) Stress-strain curves of pristine CNT fibers and the annealed fibers at different tensile tensions (0 N, 0.02 N and 0.05 N). (b) Stress-strain curves of the fibers annealed by different voltages (50 V, 60 V, 70 V and 80 V) for 12 ms under the annealing tension of 0.05 N.

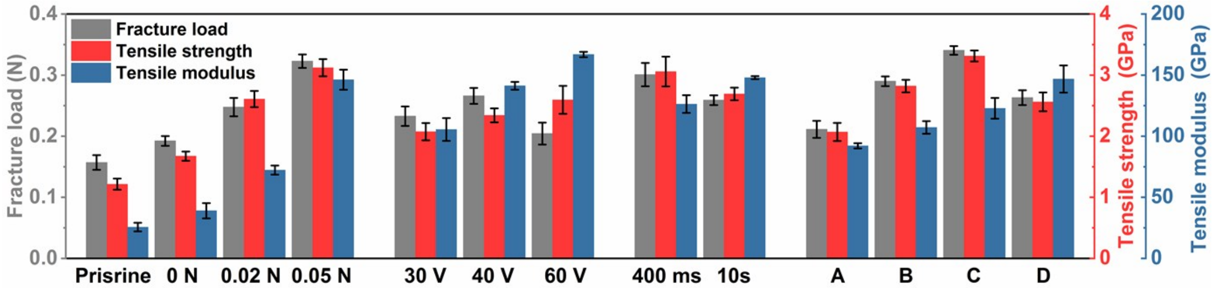

Fig.S10 Chart comparing the breaking load, tensile strength and modulus of pristine fibers and the annealed fibers fabricated under different annealing conditions.

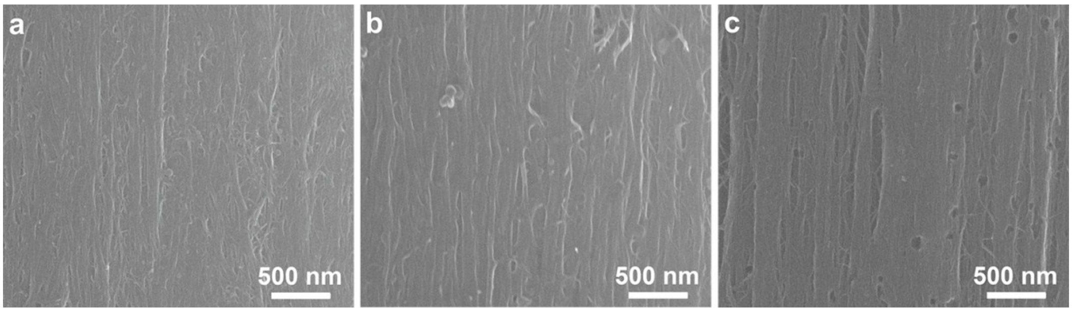

Fig.S11 (a), (b), (c) and (d) illustrate the SEM images of the fibers annealed by different voltages (30 V, 40 V and 60 V) for 5 s under the annealing tension of 0.05 N.

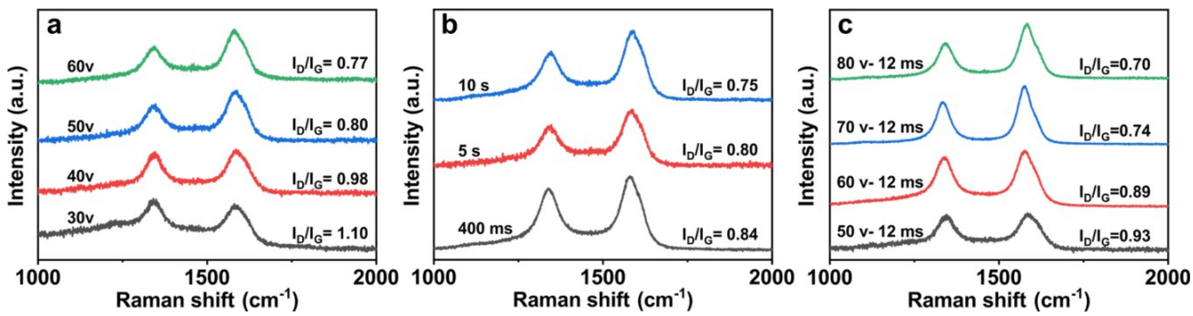

Fig.S12 (a) Raman spectras for the fibers annealed annealed by different voltages (30 V, 40 V, 50 V and 60 V) for 5 s under the annealing tension of 0.05 N. (b) Raman spectras for the fibers annealed annealed by the pulsed voltage of 50 V for different annealing times (400 ms, 5 s, and 10 s) under the annealing tension of 0.05 N. (c) Raman spectras for the fibers annealed by different voltages (50 V, 60 V,70 V and 80 V) for 12 ms under the annealing tension of 0.05 N.

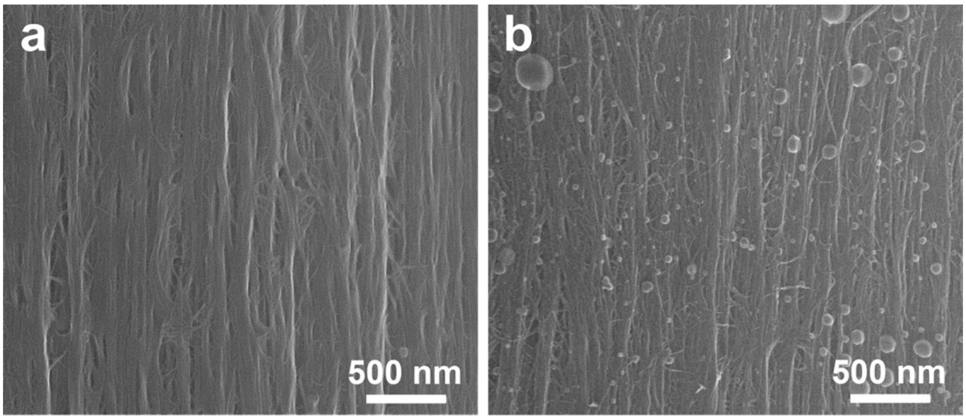

Fig.S13 (a), (b), (c) and (d) illustrate the SEM images of the fibers annealed by the pulsed voltage of 50 V for different annealing times (400 ms and 10s) under the annealing tension of 0.05 N.

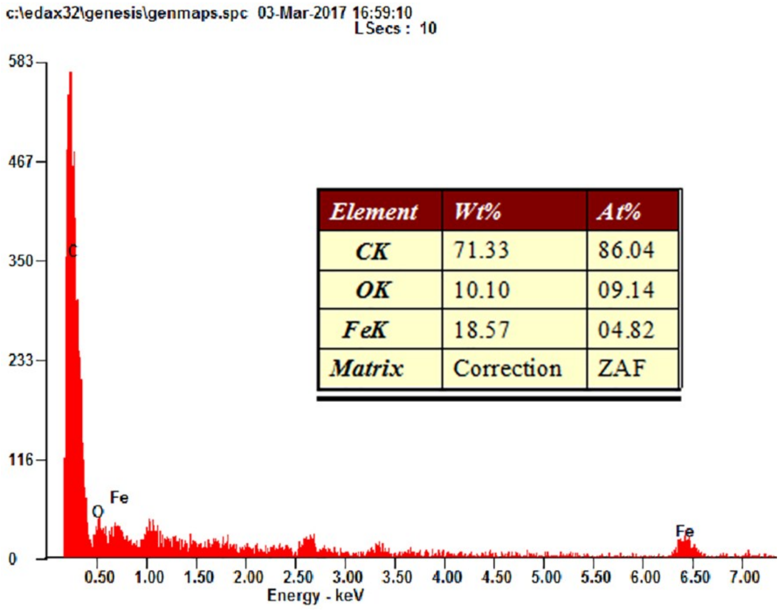

Fig.S14 Dispersive X-ray spectroscopy (EDX) analysis of nanoparticles derived by the Joule heating annealing.

When excessive annealing time and voltage were applied to the CNT fibers were, a certain amount of ablative hole defect and nanoparticles generated on the surface of CNTs and between neighboring CNTs (Figure S11c and S13b), which caused the decrease in mechanical properties. Owing to nonuniformity of the fiber resistance, the local high temperature along the fiber burned them off, leaving holes on the fiber surface [1]. The produced nanoparticles contained carbon, iron as well as oxygen elements, which was determined by means of EDX (Figure S14). It could be concluded that the generation of these nanoparticles may result from the carbon metabolism reaction involving C (fuel), O (oxidizer), and metal (catalyst).[2]

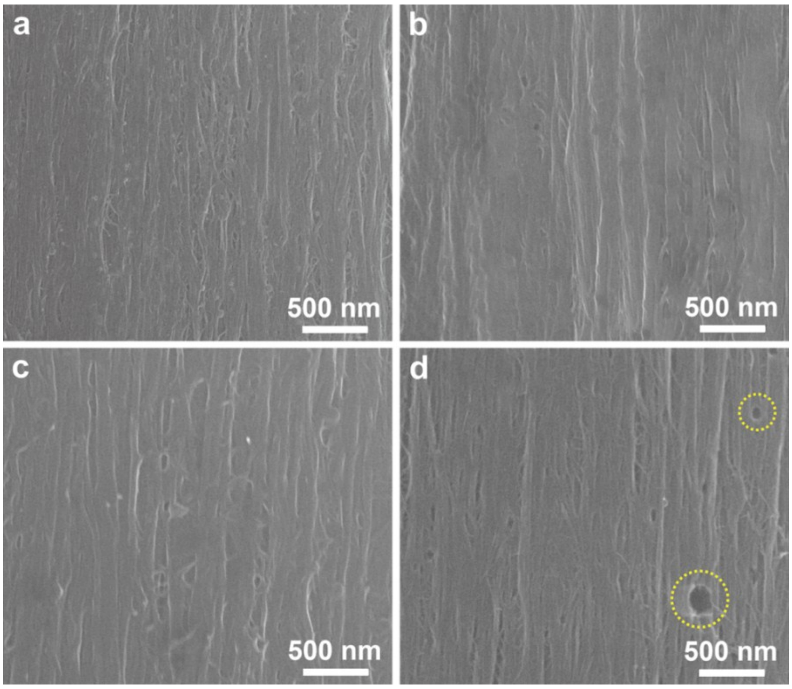

Fig.S15 (a), (b), (c) and (d) illustrate the SEM images of the fibers annealed by different voltages (50 V, 60 V, 70 V and 80 V) for 12 ms under the annealing tension of 0.05 N.

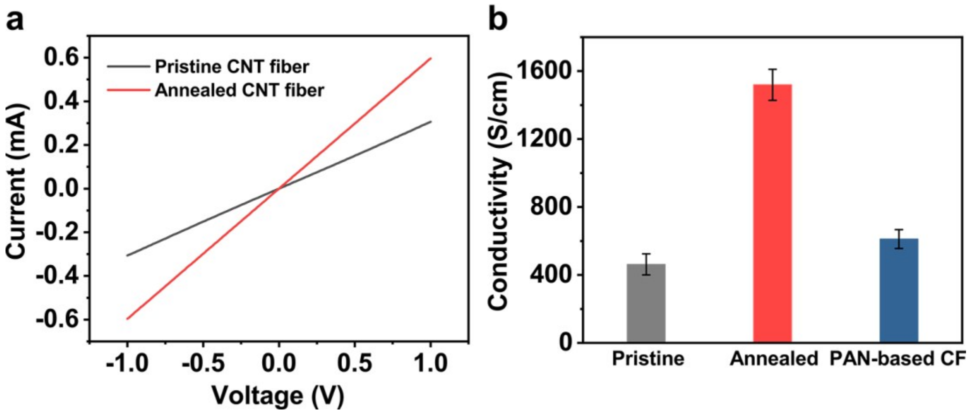

Fig.S16 (a) Conductivity measurement results for the pristine CNT fibers and the fibers annealed at a pulsed voltage of 70 V for 12 ms. (b) Chart comparing the conductivity of pristine CNT fibers, the annealed fibers and PAN-based carbon fibers.

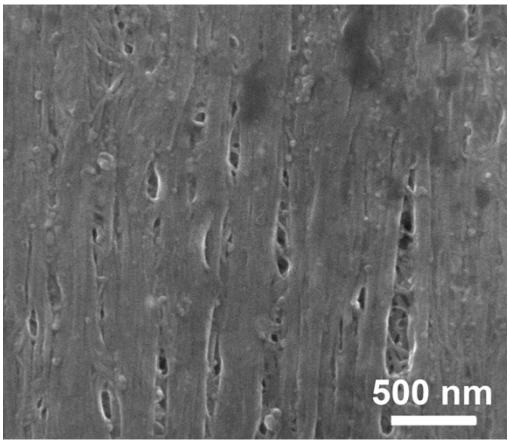

Fig.S17 SEM image of the annealed fibers after after exposure to outer flame of alcohol lamp for about 2 min.

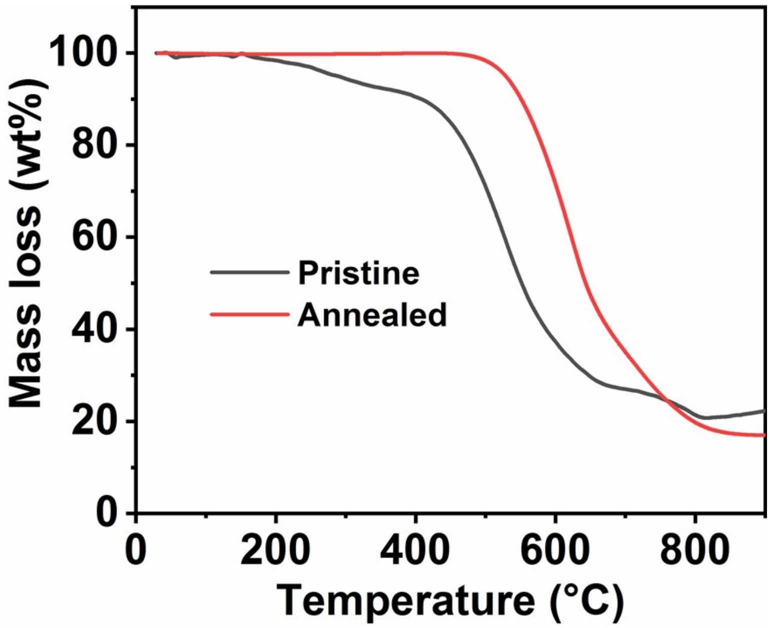

Fig.S18 TG curves of the pristine CNT fibers and the annealed fibers

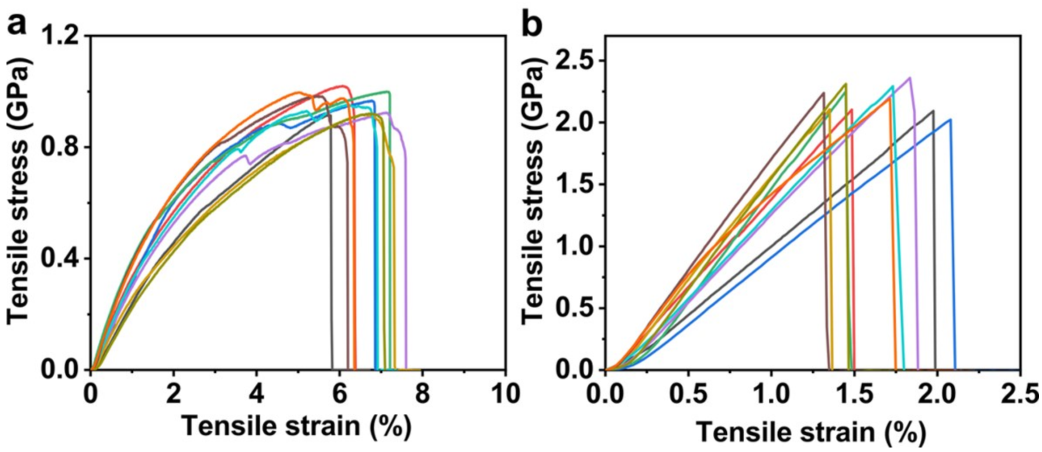

Fig.S19 (a) The stress-strains of pristine CNT fibers continuously prepared by floating catalyst CVD method. (b) The stress-strains of the annealed CNT fibers continuously fabricated by Joule heating tension-annealing method.

Video S1 showing the rapid electromechanical response properties of CNT fibers annealed by ultrafast Joule heating under tension.

Video S2 showing the changes of the formed laser diffraction fringe at power-on.

Video S3 showing ablative resistance property of the pristine CNT fibers exposed to the outer flame of alcohol lamp in air.

Video S4 showing ablative resistance property of the annealed CNT fibers exposed to the outer flame of alcohol lamp in air.

Video S5 showing continuous preparation of the annealed fibers at high-temperature Joule heating and under the applied load.

1. F. C. Meng, X. H. Zhang, R. Li, J. N. Zhao, X. H. Xuan, X. H. Wang, J. Y. Zou and Q. W. Li, *Advanced Materials*, 2014, **26**, 2480-2485.
2. Y. Yao, Z. Huang, P. Xie, S. D. Lacey, R. J. Jacob, H. Xie, F. Chen, A. Nie, T. Pu, M. Rehwoldt, D. Yu, M. R. Zachariah, C. Wang, R. Shahbazian-Yassar, J. Li and L. Hu, *Science*, 2018, **359**, 1489-1494.