One-minute Joule Annealing Enhances the Thermoelectric Properties of Carbon Nanotube Yarns *via* the Formation of Graphene at the Interface

Supporting Information

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1. Theoretical prediction of the interfacial structure of the CNTs

Note S1

Calculation of the average number of sp^2-hybridized bonds

The rate constant matrix contraction (RCMC) method[S1] classifies the local energy-minimum structures, the so-called equilibrium structures (EQs), into super states (SSs), which are defined as the states where all the EQs in each SS achieve a Boltzmann distribution for the given timescale at a given temperature. The RCMC method is a fuzzy clustering method, *i.e.*, the responsibility weight, $c_{\alpha i}$, ranges from 0 to 1. For example, if $c_{\alpha i} = 0.5$, half of the ith EQ belongs to the αth SS. The average number of sp^2-hybridized bonds in the αth SS is represented by:

$$\left\langle N^{sp^2} \right\rangle_{SS(\alpha)} = \left(\sum_{i=1}^{N_{EQ}} c_{\alpha i} e^{-\beta E_i} N_i^{sp^2} \right) \left(\sum_{k=1}^{N_{EQ}} c_{\alpha k} e^{-\beta E_k} \right)^{-1}, \quad (1)$$

where N_{EQ} is the number of EQs, β the inverse temperature, E_i the structural energy of the ith EQ, and $N_i^{sp^2}$ the number of sp^2-hybridized bonds in the structure of the ith EQ. Here, bond lengths in the range of 0.138–0.149 nm are considered sp^2-hybridized bonds. With the given initial population, the populations of the αth SS at the given time, t, $\rho_\alpha(t)$, can be calculated by the full (f)-RCMC method. Then, the average number of sp^2-hybridized bonds at time t is:

$$\left\langle N^{sp^2} \right\rangle(t) = \sum_{\alpha=1}^{N_{ss}} \rho_\alpha(t) \left\langle N^{sp^2} \right\rangle_{SS(\alpha)}. \quad (2)$$

In this study, we have assumed that the population of the initial structure was unity and all others were zero.

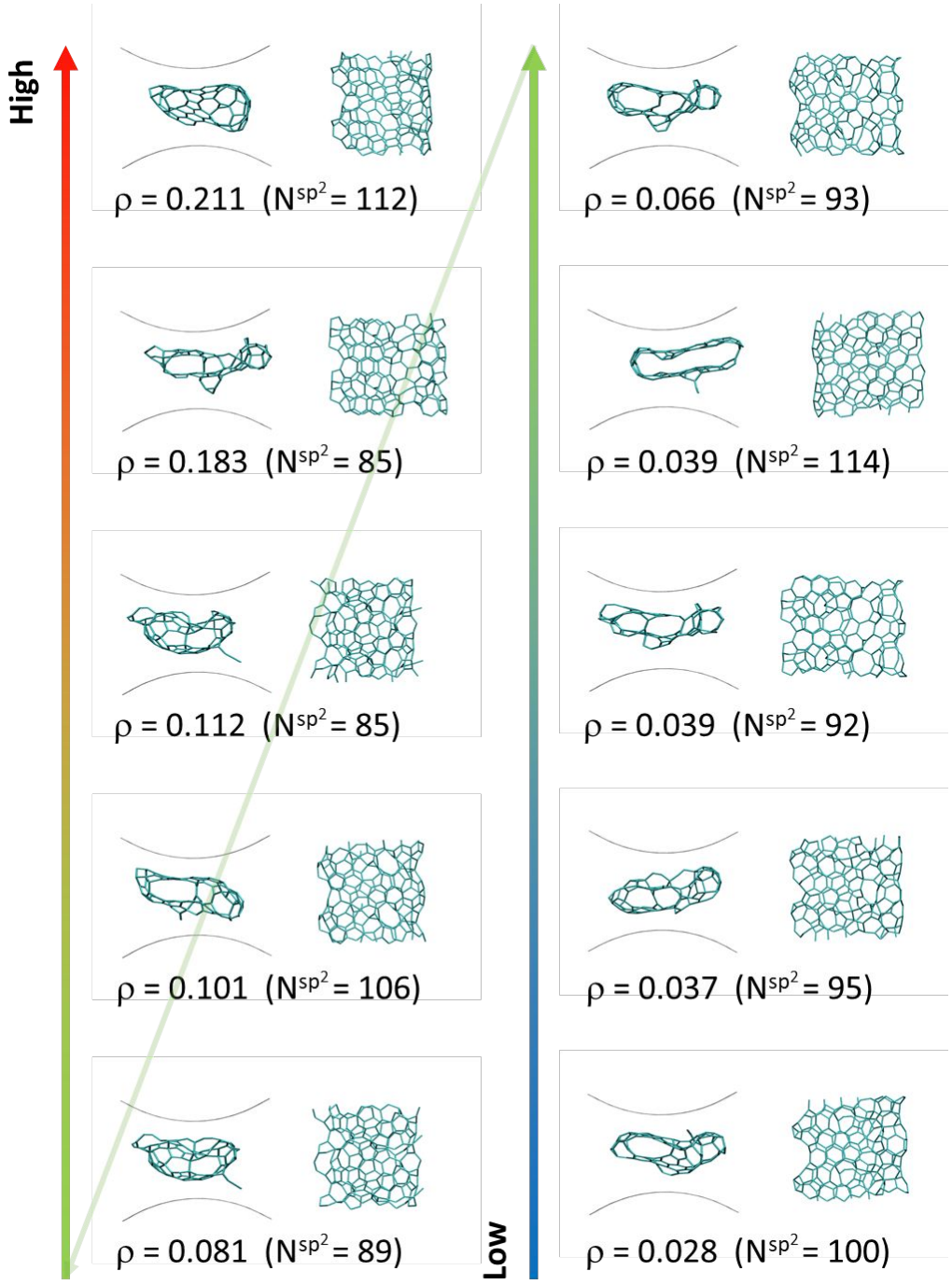

Figure S1. Top 10 most populated local energy-minimum structures after one-minute annealing at 2000 K predicted by the f-RCMC method. CNTs are shown in line representation, while amorphous carbon atoms are shown in stick representation. The right figures in each column are the top views, while the left figures are the cut views. The unit cell is doubled along the axial direction of the CNTs for clarity. The number of sp^2-hybridized bonds is shown in parentheses. The molecular graphics were created using the Visual Molecular Dynamics (VMD) program[S2].

2. Fabrication and characterization of the CNT yarns

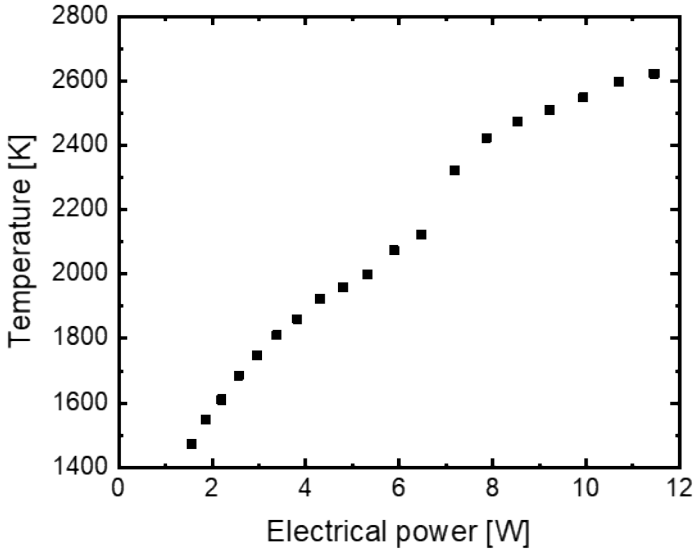

Figure S2. The annealing temperature of a CNT yarn as a function of the applied electrical power. The temperature was measured using a radiation thermometer. The typical applied power was 5 W; therefore, the temperature of the CNT yarn during annealing was ~ 2000 K.

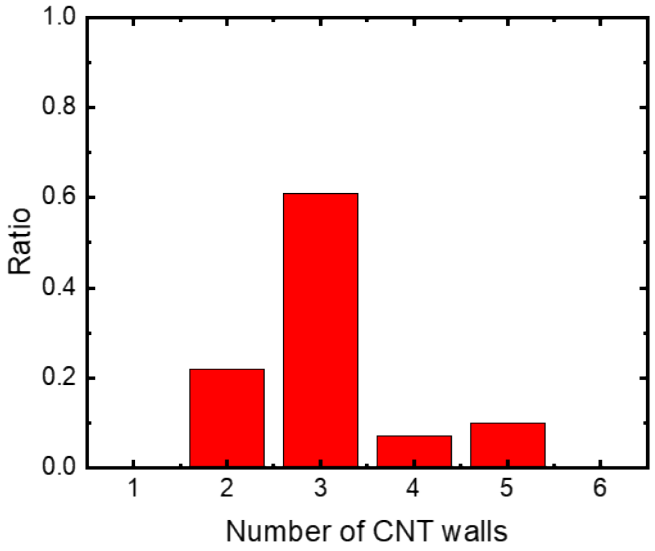

Figure S3. The number of CNT walls obtained from TEM images. One hundred CNTs were evaluated.

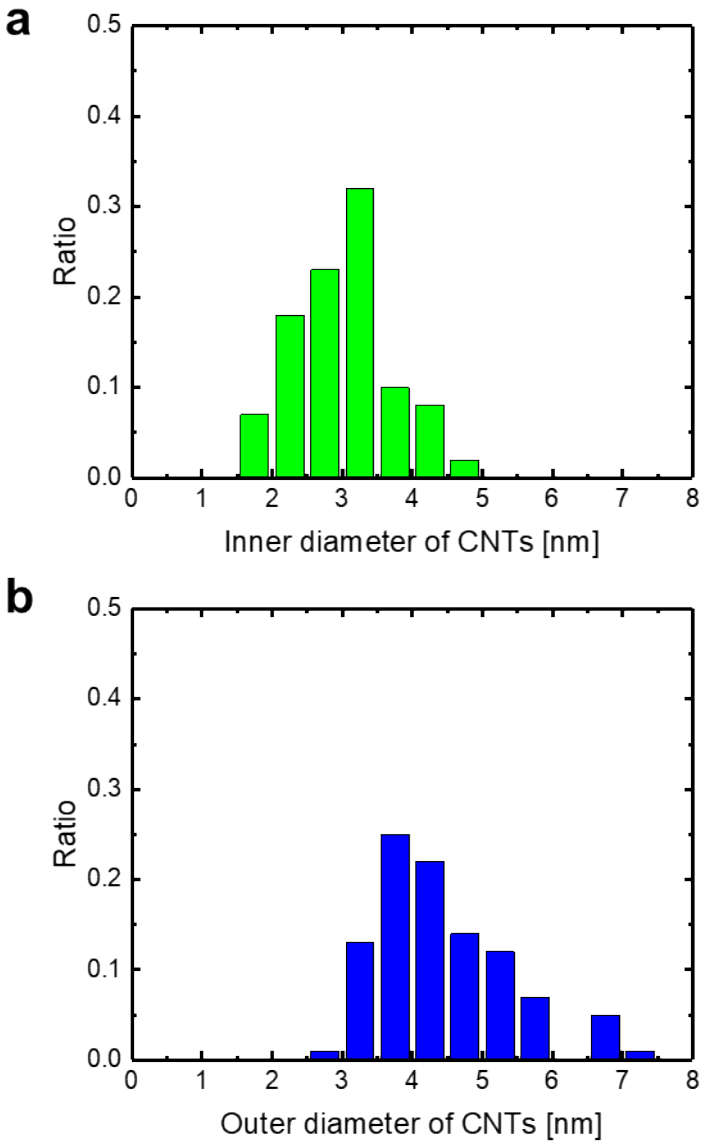

Figure S4. Inner (a) and outer (b) diameter of the CNTs obtained from TEM images. One hundred CNTs were evaluated.

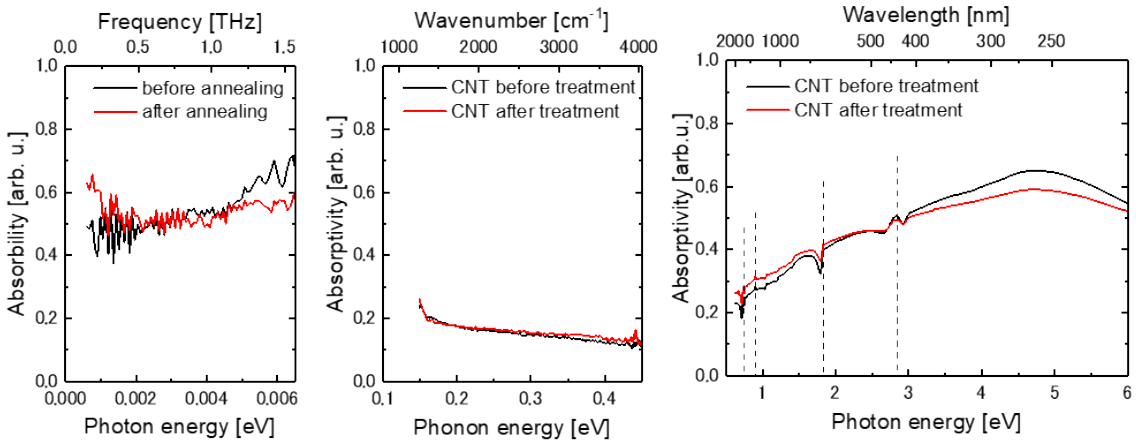

Figure S5. Absorption spectra. Optical transmission spectra of the CNT webs in the terahertz, mid-IR, visible, and ultraviolet regions. The absorption spectra did not change significantly after annealing.

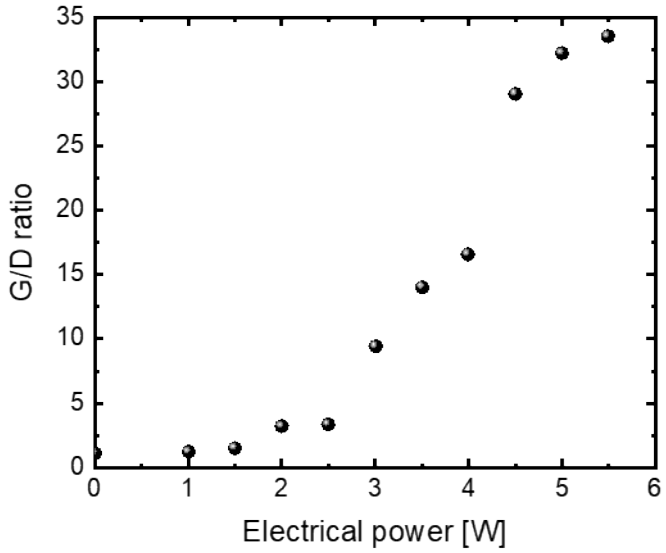

Figure S6. Raman G/D ratio of annealed CNT yarns as a function of the electrical power.

3. Thermoelectric and mechanical properties of the CNT yarns

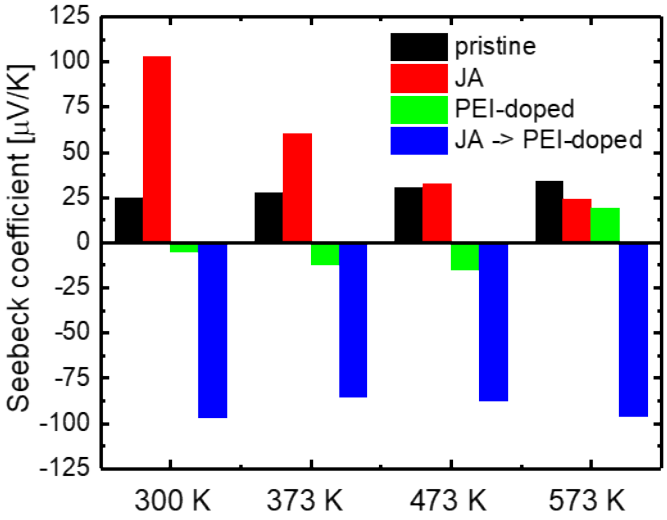

Figure S7. Temperature dependence of the Seebeck coefficients of the pristine, Joule-annealed (JA), PEI-doped, as well as PEI-doped and Joule-annealed CNT yarns.

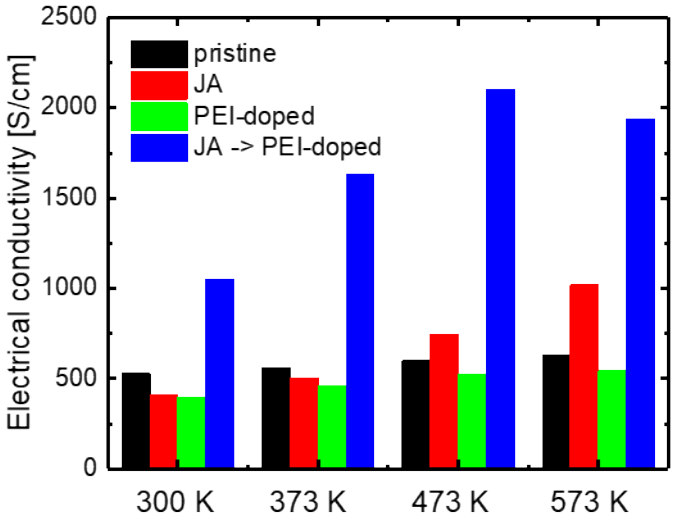

Figure S8. Temperature dependence of the electrical conductivity of the pristine, Joule-annealed (JA), PEI-doped, as well as PEI-doped and Joule-annealed CNT yarns.

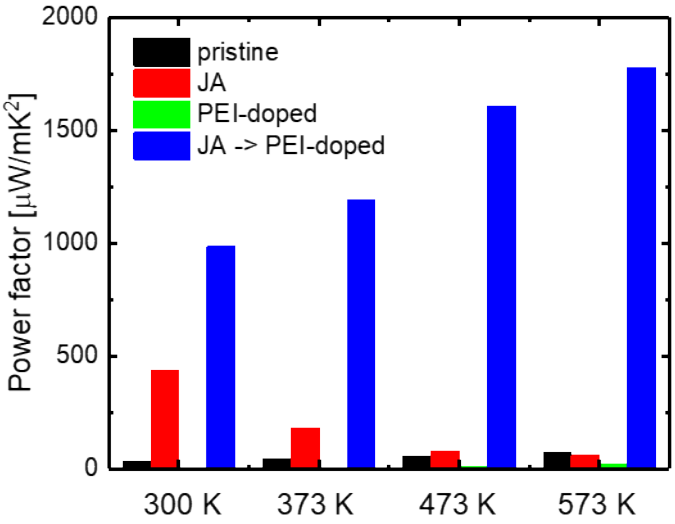

Figure S9. Temperature dependence of the thermoelectric power factors of the pristine, Joule-annealed (JA), PEI-doped, as well as PEI-doped and Joule-annealed CNT yarns.

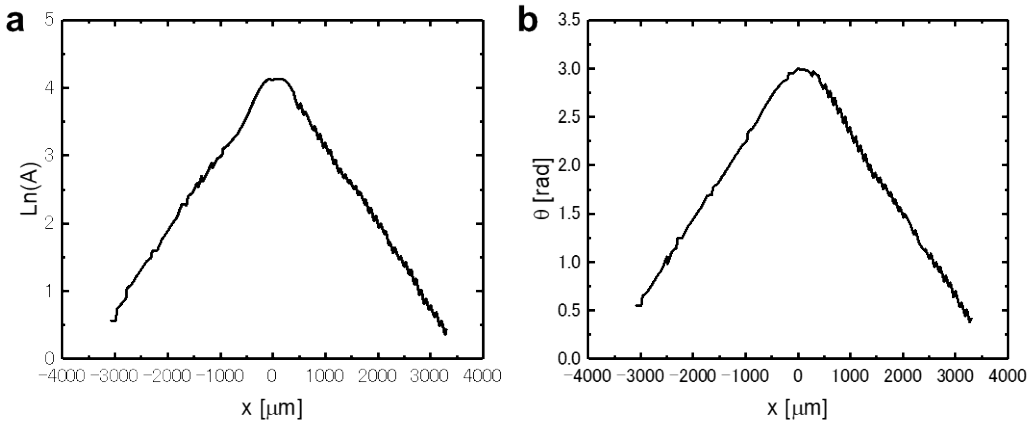

Figure S10. The thermal diffusivity of the CNT yarns measured by modified AC calorimetry.[S3] The thermal diffusivity from amplitude (α_a) (a) and phase (α_p) (b) are given by $A \propto \exp\left(-x\sqrt{\omega/2\alpha_a}\right)$ and $\theta \propto -x\sqrt{\omega/2\alpha_p}$, where A and θ the amplitude and phase of temperature-modulation measured by a thermocouple, x, and ω are the heated position from the thermocouple and angular frequency ($\omega = 2\pi f$, f = 10 Hz). The thermal diffusivity (α) is given by $\alpha = \sqrt{\alpha_a \cdot \alpha_p}$. The thermal diffusivity of the CNT yarns was measured to be 3.27 ± 0.01 × 10^{-5} m^2/s. Thermal conductivity (k) was calculated with the density (ρ) and specific heat (c) of the material as $\alpha = k/\rho c$. As mentioned in the main text, density and specific heat of the material are 1.0 g/cm^3 and 0.74 J/g·K, respectively.[S4]

Supplementary References

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