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Supplementary Information

Optimization of Thermoelectric Properties of Carbon Nanotube Veils by Defect Engineering

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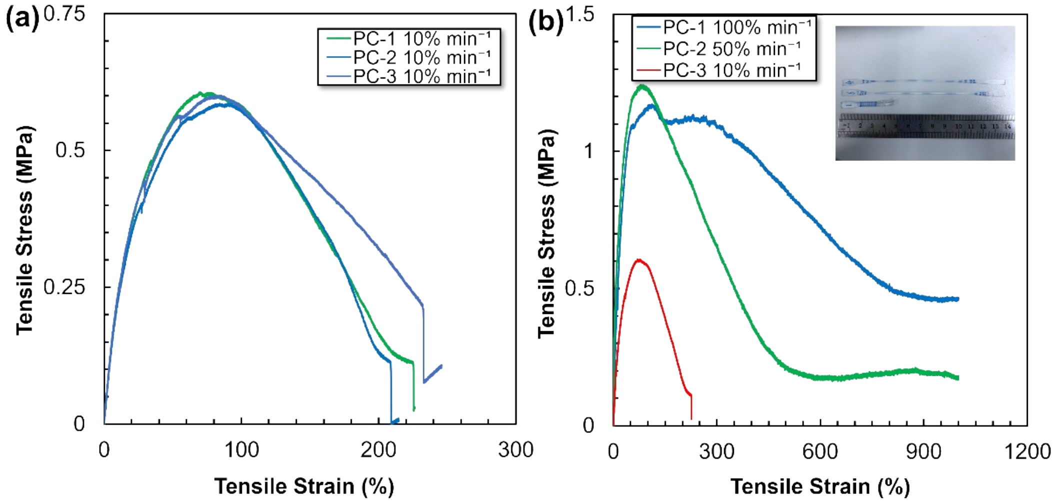

Figure S1. Mechanical properties of PC films at 160 °C with load speed of 10% min^{-1} (a) and other speed (b). Stretched PC films with different load speed are inset in (b).

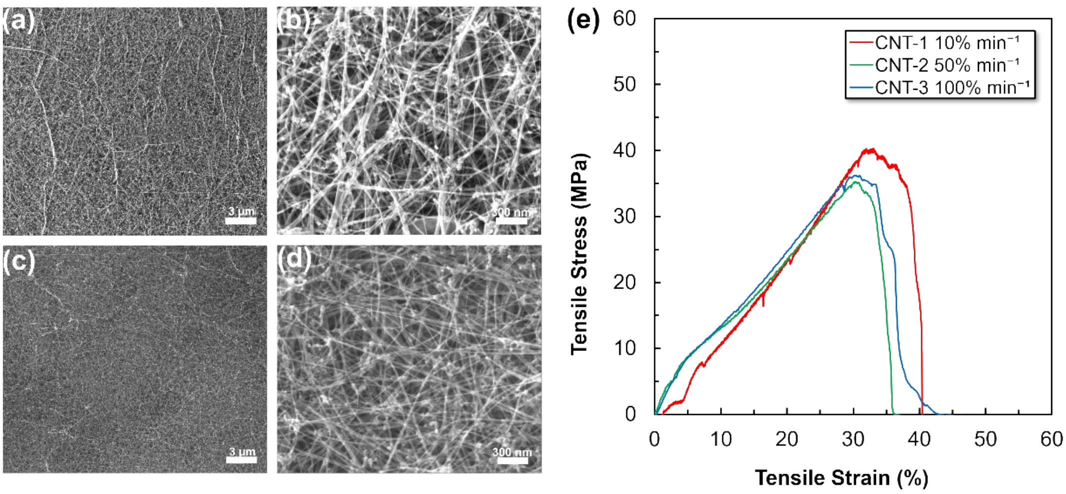

Figure S2. SEM images of CNT veils (a-d), surface (a-b) and bottom (c-d). Mechanical properties of CNT veils at 160 °C with different load speed (e). Scale bars are 3 µm and 300 nm, respectively.

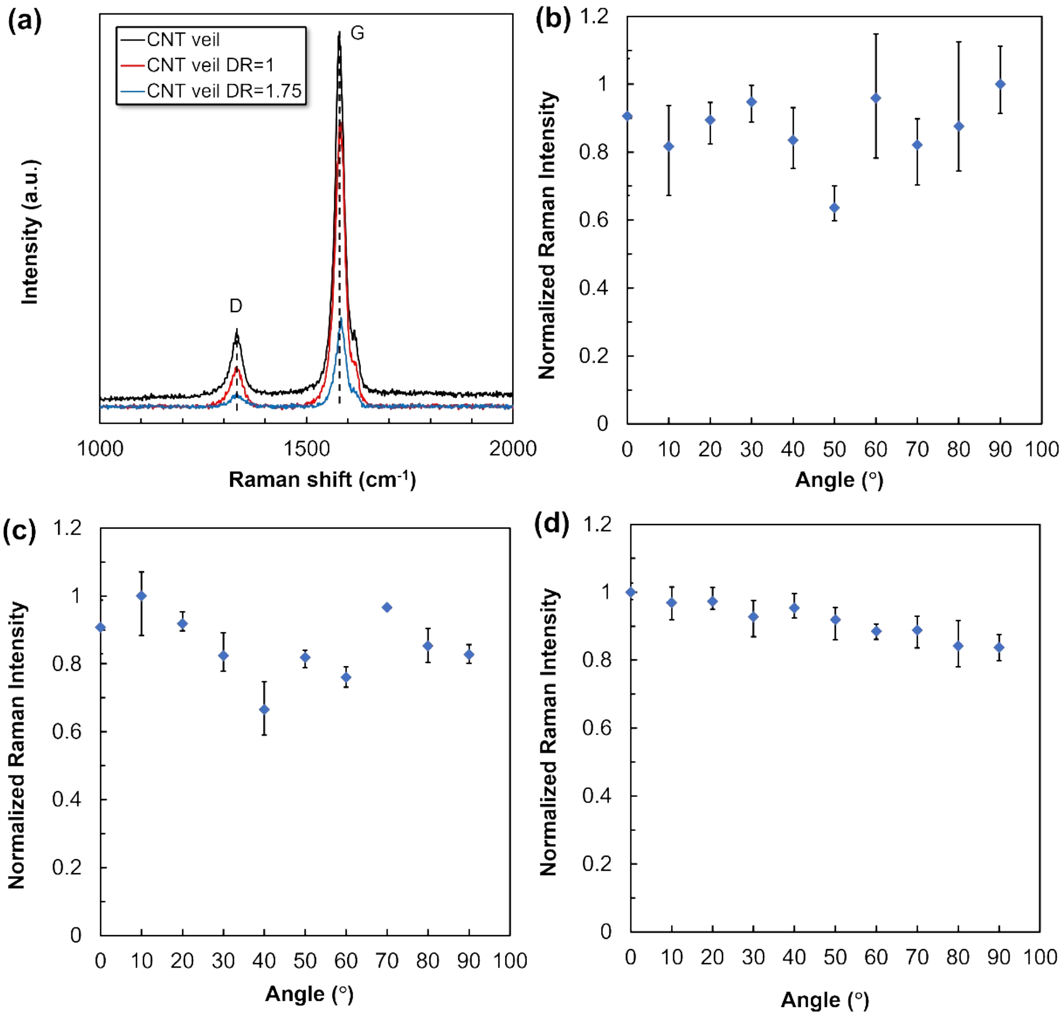

Figure S3. Raman spectra of CNT veils excited with a 633 nm laser (a) and normalized Raman intensity of the G-band for: pure CNT veil (b), PC/CNT/PC of DR = 1 after dissolving PC layer (c) and stretched PC/CNT/PC of DR =1.75 after dissolving PC layer (d).

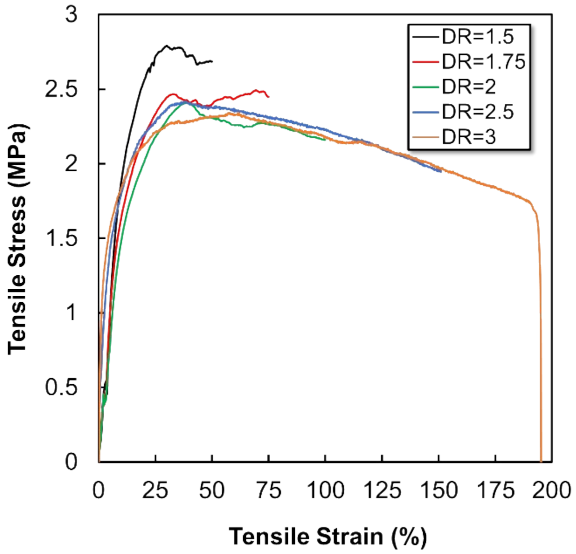

Figure S4. Mechanical properties of PC/CNT/PC with different draw ratios.

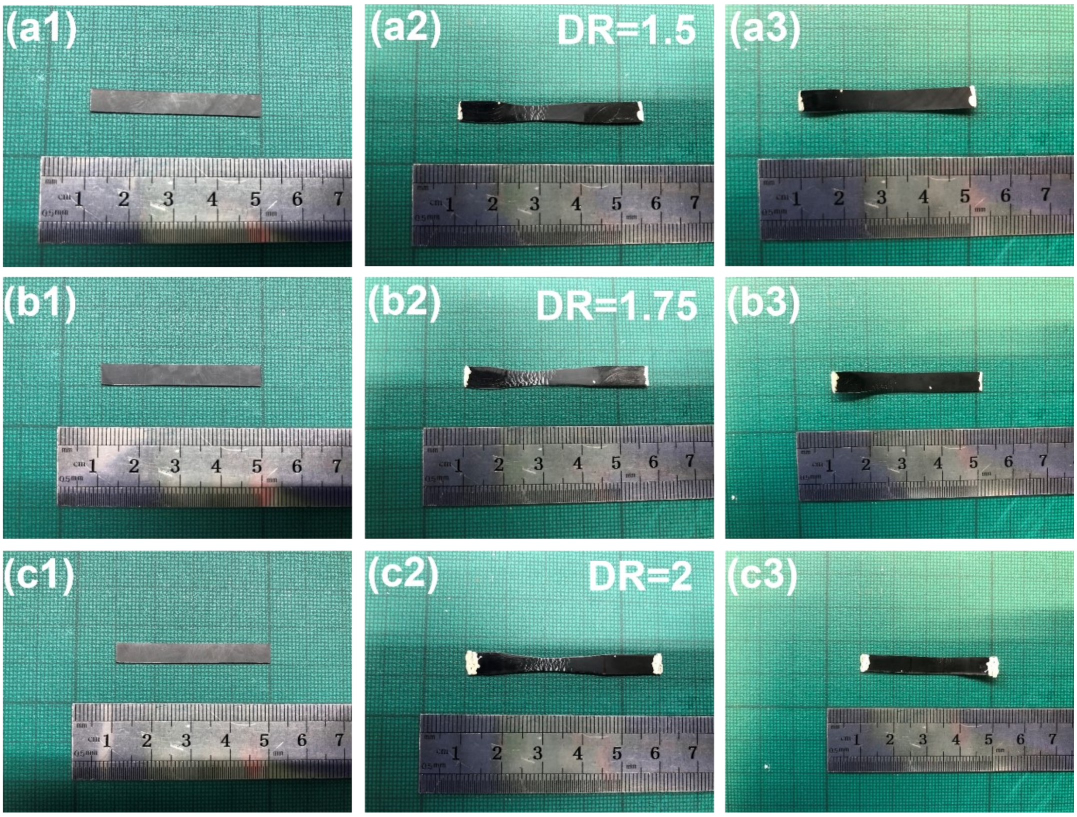

Figure S5. Digital photos of PC/CNT/PC composites at initial, stretched and heat-repaired states with different DR. DR = 1.5 (a1-a3), 1.75 (b1-b3) and 2 (c1-c3).

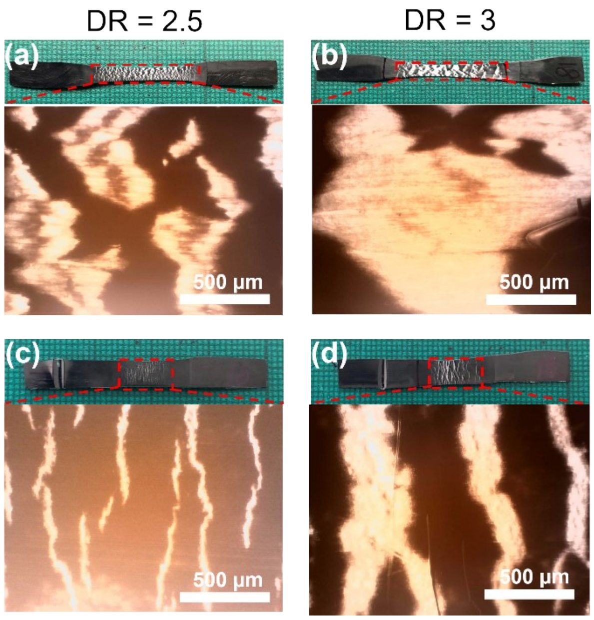

Figure S6. Digital photos and microscope images of stretched (a-b) and heat-repaired PC/CNT/PC composites (c-d) with DR of 2.5 and 3. Scale bar is 500 μm.

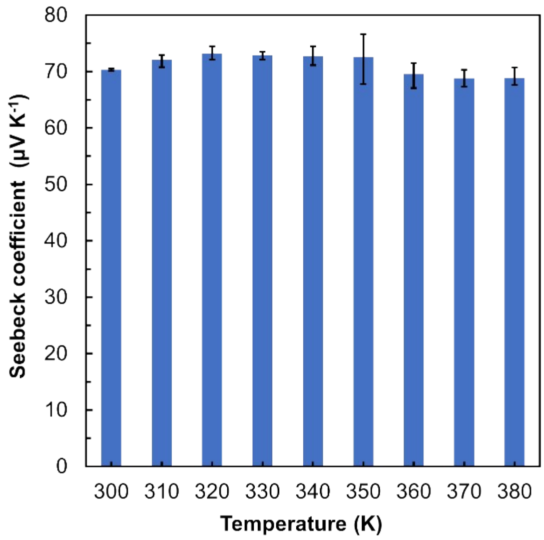

Figure S7 The dependence of the Seebeck coefficient of CNT veils on temperature.

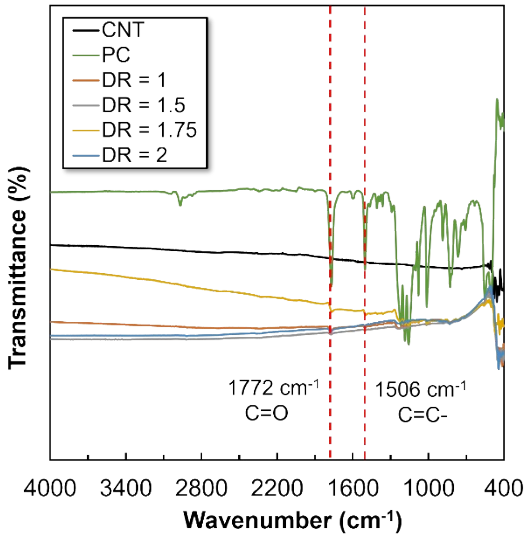

Figure S8. FTIR spectra of CNT veil, PC film and CNT veils after dissolving PC layer.

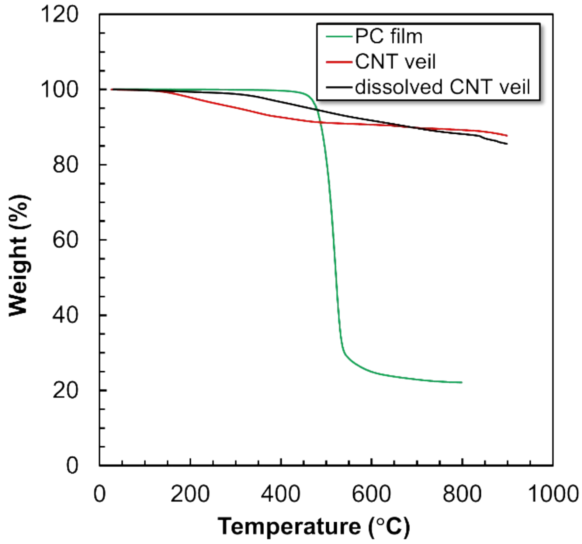

Figure S9. TGA results of PC film, CNT veil and CNT veil after dissolving PC layer.

Note S1. Calculation the interfacial shear strength

The apparent shear strength at the fiber-matrix interface (τ) is given as follows:[1]

$$\tau = \frac{\sigma_f d}{2l_c} \tag{1}$$

From this equation, σ_f is the ultimate tensile strength of the fiber, d is the diameter of the fiber, l_c is the critical fiber length. In our case, CNT veil in the PC/CNT/PC composites is treated analogously to a continuous fiber. Hence, the interfacial shear strength can be estimated by this method. Here the force due to shear at the matrix/CNT veil interface and the force carried by the CNT veil are as follows,

$$F_1 = \tau\frac{l_c}{2}(2t + 2w) \tag{2}$$

$$F_2 = \sigma_{CNT} wt \tag{3}$$

Equating (2) and (3), we can obtain,

$$\tau \approx \frac{\sigma_{CNT} t}{l_c} \tag{4}$$

where t is the thickness of CNT veil, σ_{CNT} is the ultimate tensile strength of CNT veil, l_c is the critical length of CNT veil. According to the results of Fig. S2 and Fig. 3, the average σ_{CNT} and mean l_c are 37.2 MPa and 310 μm, respectively and t is 3.6 μm. Based on the relationship between critical length and the interfacial shear strength (Equation 4), the interfacial shear strength between the CNT veil and PC can be calculated as 0.43 MPa.

Note S2-Calculation of thermal conductivity contributions

The in-plane electrical thermal conductivity for CNT film can be estimated on the basis of the Wiedemann-Franz law:[2]

$$\kappa_e = L_0 \times \sigma \times T \tag{5}$$

From this equation, κ_e is the electronic thermal conductivity, L_0 is the Lorenz number of 2.44 × 10^{-8} W Ω^{-1} K^{-2} (the Lorenz number dependent on materials and may not be applicable to the samples in this work), σ is the electrical conductivity of CNT veil and T is the temperature. Based on the equation, the calculated κ_e is 0.64 W m^{-1} K^{-1}. Compare to the κ of the CNT veil (46 W m^{-1} K^{-1}), the ratio of the electron to phonon contribution to the thermal conductivity is about 1 to 71, which suggests that the heat transport is dominated by the phonon component in the CNT veil.

Table S1. Hall measurements of the CNT veil

Material	Hall coefficient ($cm^3\ C^{-1}$)	Carrier density (cm^{-3})	Carrier mobility ($cm^2\ V^{-1}\ s^{-1}$)
CNT veil	2.27×10^{-3}	3.17×10^{21}	1.89

Table S2. Raw data and normalized data of six performance indexes for CNT veil and composites.

Name	CNT veil after removing PC layer		Composites			
			Parallel model		Series model	
DR	1	1.5	1	1.5	1	1.5
σ (S cm^{-1})	289	214	289	214	289	214
S (μV K^{-1})	55	54	55	54	55	54
κ (W m^{-1} K^{-1})	46	13	1.63	0.68	0.34	0.34
1/κ (m K W^{-1})	0.0216	0.0778	0.6119	1.4608	2.9662	2.9678
ZT	0.0006	0.0015	0.0158	0.0273	0.0768	0.0554
E (MPa)	144	/	745	854	745	854
Max tensile stress (MPa)	31.9	/	46.7	44.8	46.7	44.8

Normalized data (each physical property is divided by the maximum value achieved within this study – underlined above – to which the arbitrary value of 10 is assigned).

Name	CNT veil after removing PC layer		Composites			
			Parallel model		Series model	
DR	1	1.5	1	1.5	1	1.5
σ	10	7.4	10	7.4	10	7.4
S	10	9.87	10	9.87	10	9.87
1/κ	0.0729	0.2620	2.06	4.92	10	10
ZT	0.0730	0.1892	2.06	3.55	10	7.22
E	1.68	/	8.72	10	8.72	10

Max tensile stress (MPa)	6.83	/	10	9.59	10	9.59

References

1. O. Tadashi, A. Nakayama, M. Miwa and A. Hasegawa, *J. Appl. Polym. Sci.,* 1978, **22**, 3203-3212.
2. W.B. Zhou, Q.X. Fan, Q. Zhang, L. Cai, K. W. Li, X. G. Gu, F. Yang, N. Zhang, Y.C. Wang, H.P. Liu, W.Y. Zhou, S.S Xie, *Nat. commun.*, 2017, **8**, 14886.