

Supplementary Information for

Toward improved trade-off between thermoelectric and mechanical performances in polycarbonate/single-walled carbon nanotube composite films

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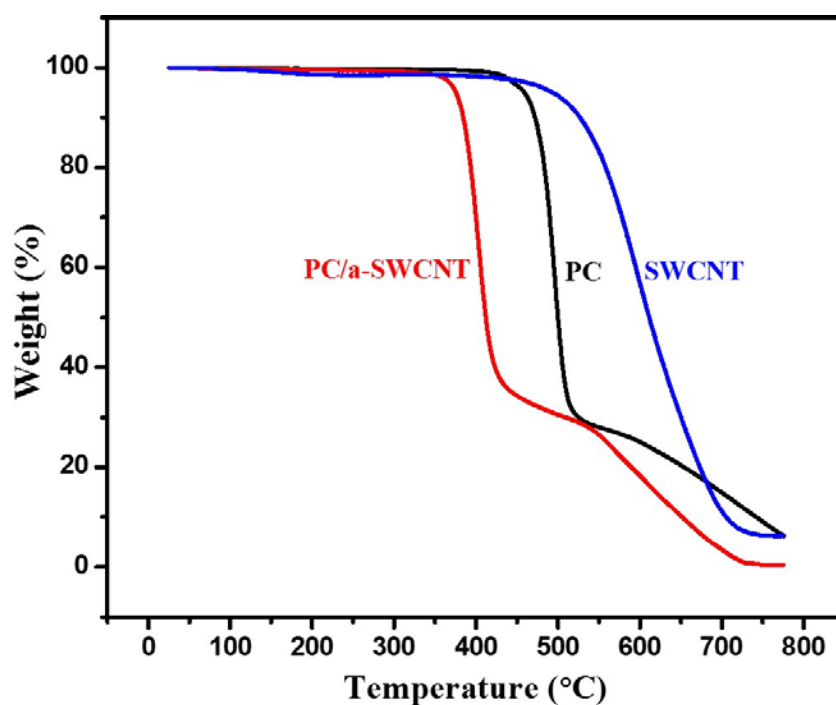
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Supplementary I. Thermogravimetric Analysis

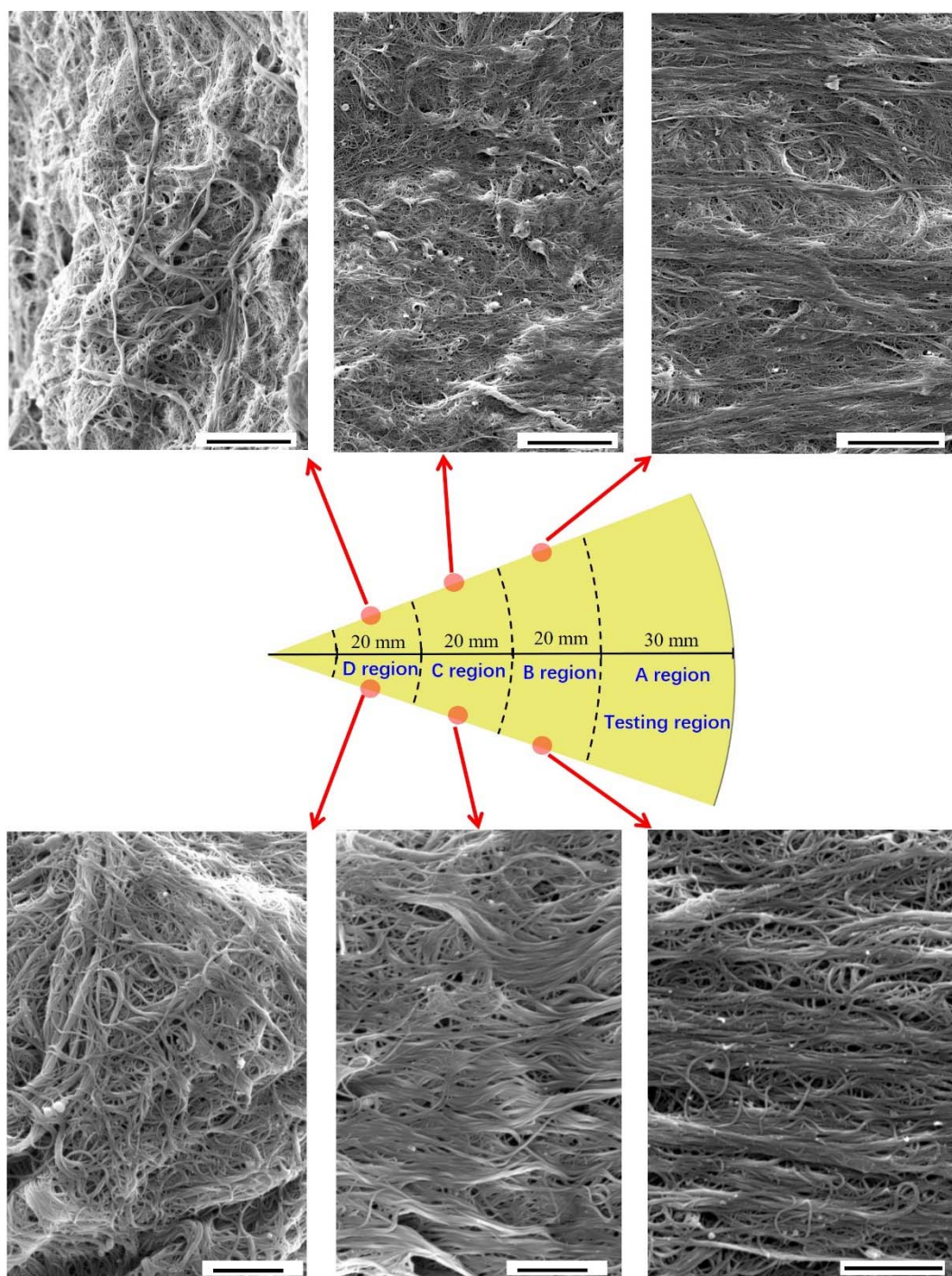
Thermogravimetric Analysis (TGA) was carried out to study the thermal decomposition behaviors of PC/a-SWCNT composite, neat PC and SWCNT in Supplementary Fig. 1. The TGA curves of neat PC and SWCNT show that the onset degradation/ decomposition temperatures for neat PC and SWCNT are 457.9 °C and 495.3 °C, respectively. The PC/a-SWCNT composite starts to degrade at 375.8 °C, much higher than our melt-processing temperature (280 °C). Therefore, no distinct degrade occurs in the melt-processing of the composite films.



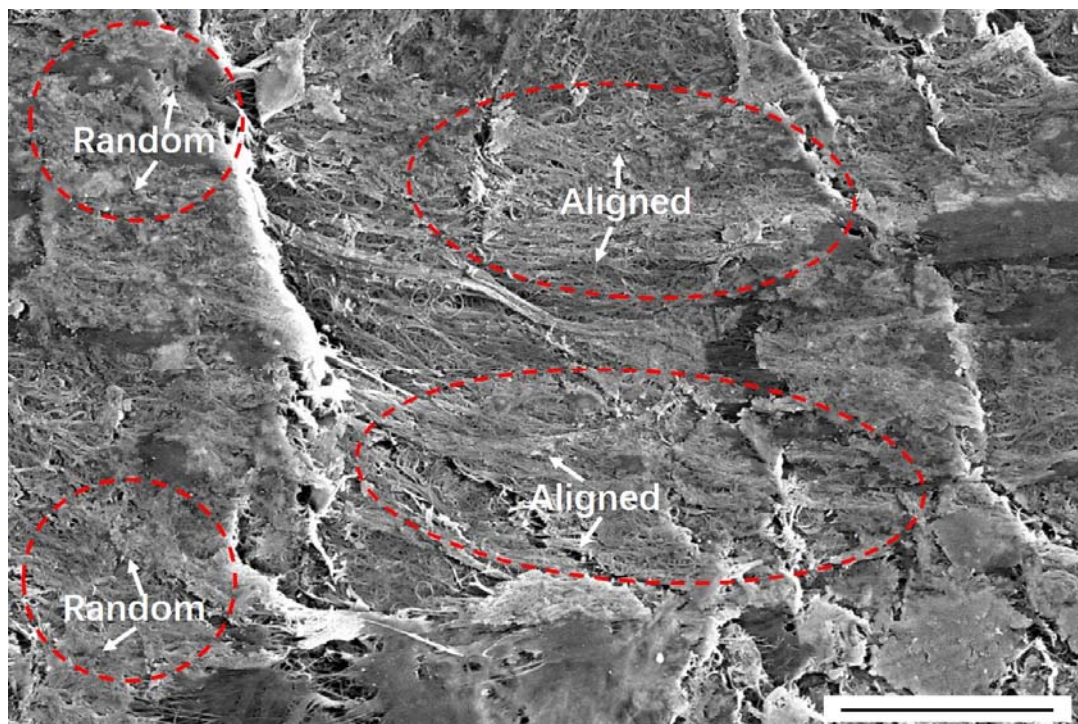
Supplementary Fig. 1 TGA curves of PC/a-SWCNT composite with 4 wt% of SWCNT, neat PC and SWCNT.

Supplementary II. SWCNT alignment along the radial direction

In order to check the dispersed states of SWCNTs along the radial direction, we observed different positions away from the edge of the nearly circular sample. The samples are divided into four regions, the approximate middle positions of which are characterized and presented in Supplementary Fig. 2. SEM images display the random SWCNT dispersed states in the form of fine bundles with big sizes in “D region”. By contrast, it presents obvious changes of the SWCNT dispersed states in “C region”. Many aligned bundles of SWCNTs appear in “C region”. It is supposed that many random SWCNTs transform to the aligned SWCNTs in this region, as also indicated in Supplementary Fig. 3. As expected, “B region” shows quite fine alignment of SWCNTs. The fine alignment of SWCNTs is also achieved in “A region” that is the edge of the nearly circular sample, as confirmed in Fig. 1D and 1F. The increasing squeeze flow should be responsible for the aligned SWCNTs along the radial direction.¹ Similar phenomena on the compression-induced fiber orientation were also reported by Park et al.,² who numerically and experimentally found fiber orientation in compression-molded polymer composites. The samples for thermoelectric and mechanical tests are from “A region” to ensure high degree of alignment of SWCNTs.



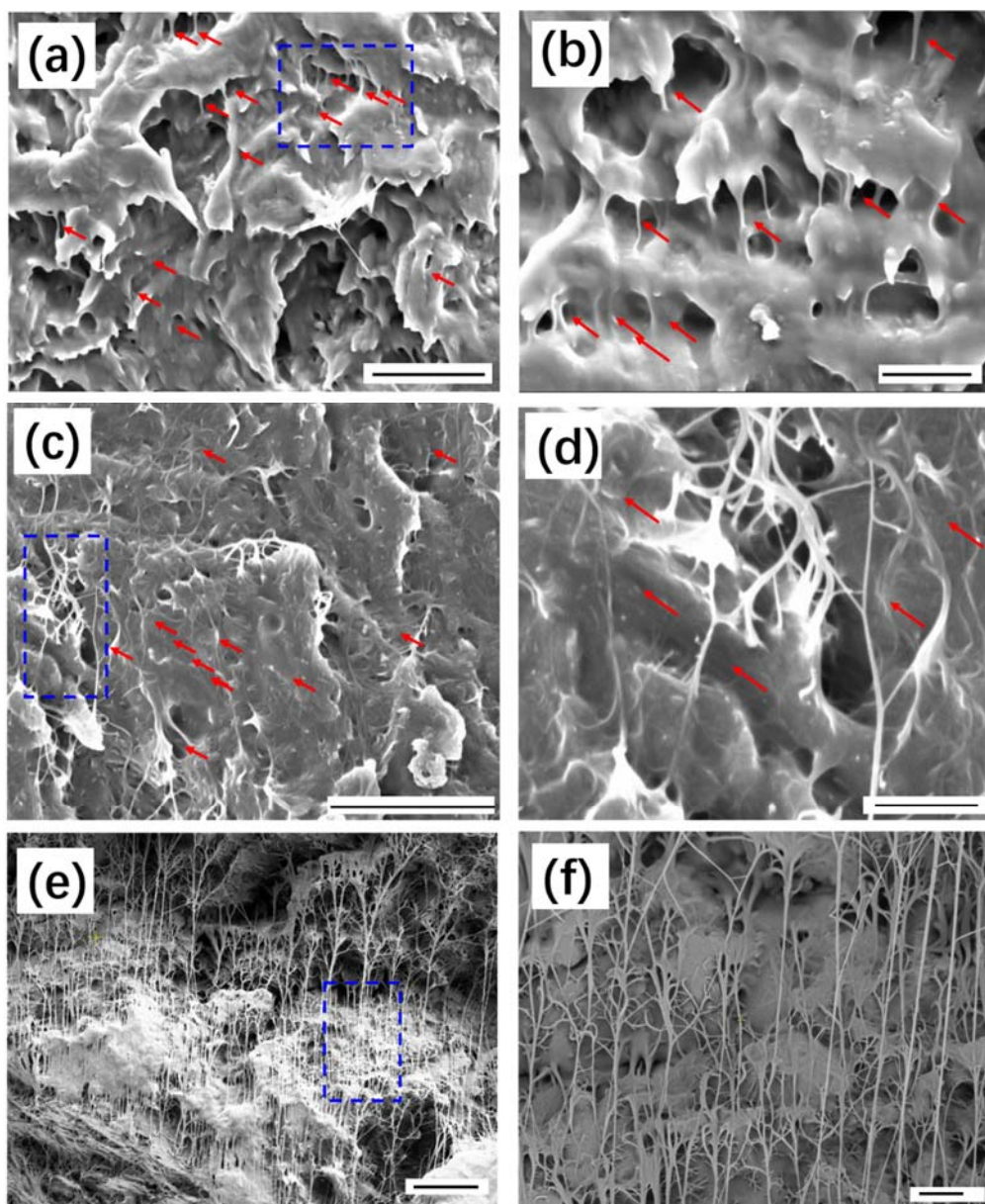
Supplementary Fig. 2 SEM images of the PC/a-SWCNT composite film at A–D regions according to the distance away from the film edge, where the upper and the bottom correspond to before and after being etched by CH_2Cl_2 . The SWCNT content is 10 wt%. The scale bars in upper 3 SEM images and bottom 3 SEM images are 5 and 1 μm , respectively.



Supplementary Fig. 3 SEM image of “C region” of PC/a-SWCNT composite film.

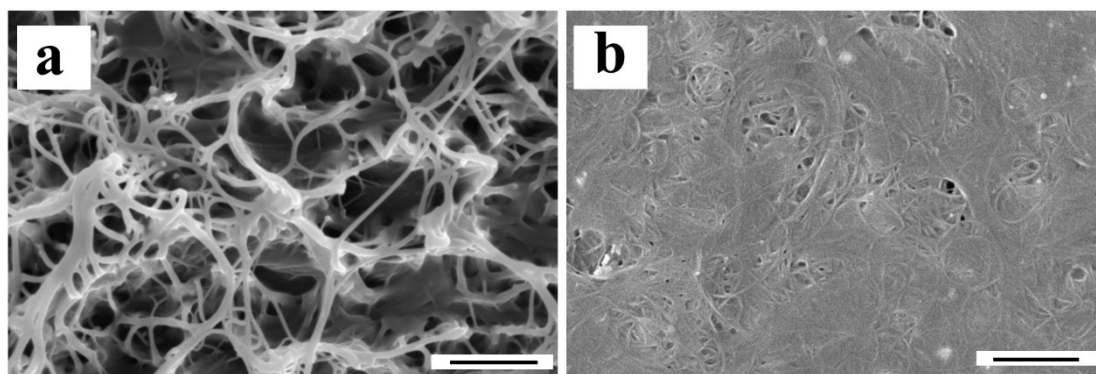
The scale bar is 5 μm .

Supplementary III. SEM images of PC/a-SWCNT composite films with other SWCNT contents



Supplementary Fig. 4 SEM images of cross sections of PC/a-SWCNT composite films at SWCNT contents of (a, b) 4 wt%, (c, d) 6 wt% and (e, f) 8 wt%. The SEM images on the right side are from enlarged areas of their left sides marked by blue color. The scale bars in left 3 SEM images and right 3 SEM images are 5 and 1 μm , respectively.

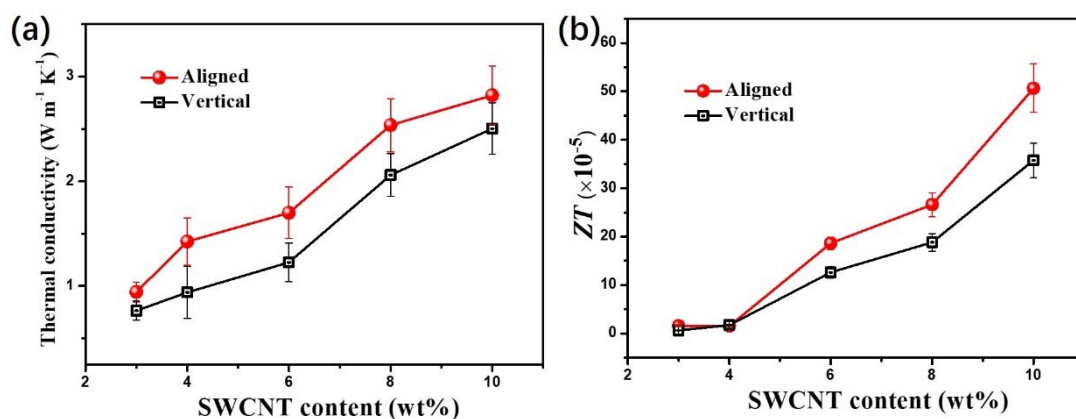
Supplementary IV. The SWCNT dispersion in PC/r-SWCNT film



Supplementary Fig. 5 High-magnification SEM images of Fig. 1E for PC/r-SWCNT composite films. The scale bar is 1 μm .

Supplementary V. Thermal conductivity and ZT values for PC/a-SWCNT films

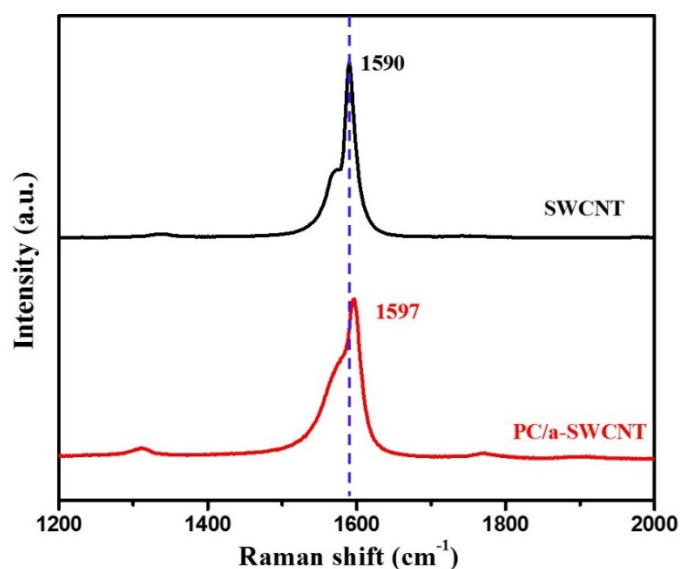
The parallel thermal conductance method was performed to measure the thermal conductivities of PC/a-SWCNT films in the aligned and vertical directions.³ The detailed description about the apparatus was reported in previous works.^{4,5} Supplementary Fig. 6a suggests that the SWCNT alignment causes a slightly higher thermal conductivity of PC/a-SWCNT films in the aligned direction. Combining thermal conductivity and power factor, the values of TE figure of merit (defined as $ZT = S^2\sigma T/\kappa$, where T and κ are the absolute temperature and the thermal conductivity, respectively) are compared for PC/a-SWCNT composites at both directions in Supplementary Fig. 6b. Apparently, the PC/a-SWCNT composite shows an anisotropic TE performance with a higher ZT value in the aligned direction.



Supplementary Fig. 6 The thermal conductivities (a) and ZT values (b) of PC/a-SWCNT films in the SWCNT direction and vertical directions. Error bars represent standard deviation with $n \geq 3$ for each measurement.

Supplementary VI. Raman spectra of SWCNT and PC/a-SWCNT

Raman analysis was carried out to reveal the information on interfacial interaction between the PC and SWCNTs. Supplementary Fig. 7 shows a typical G band at 1590 cm^{-1} that is assigned to the E_{2g} mode related to the vibration of sp^2 -bonded carbon atoms in 2D hexagonal lattices.⁶ This strong G-band peak indicates a large quantity of conjugated carbon structures in the SWCNTs.⁷ For the PC/SWCNT composite, the strong peak for the G band peak is still observed, distinctly shifting to 1597 cm^{-1} . The band-shift confirms the strong interfacial interaction between PC chains and SWCNTs, which results in effective stress transfers in the composite films.^{8,9}



Supplementary Fig. 7 Raman spectra for the SWCNT and PC/a-SWCNT composite with 4 wt% of SWCNTs.

Supplementary VII. Table of the mechanical tensile properties

Supplementary Table 1. Mechanical tensile properties for the PC/a-SWCNT composite films as a function of SWCNT content. Error bars represent standard deviation with $n \geq 5$ for each measurement.

Contents of Samples	Tensile modulus (GPa)	Fracture strength (MPa)	Elongation at break (%)
0 (Neat PC)	1.0 ± 0.1	57.5 ± 6.1	81.3 ± 10.9
3 wt%	1.4 ± 0.3	84.4 ± 7.8	10.5 ± 2.3
4 wt%	1.7 ± 0.2	74.8 ± 7.8	6.4 ± 1.2
6 wt%	2.1 ± 0.2	53.9 ± 8.6	2.8 ± 0.8
8 wt%	2.7 ± 0.2	46.6 ± 6.1	2.4 ± 0.9
10 wt%	3.1 ± 0.3	44.1 ± 8.9	2.1 ± 0.6

Supplementary References

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