Revealing the Essential Effect Mechanism of Carbon Nanotubes on The Thermal Conductivity of Graphene Film

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Supporting information

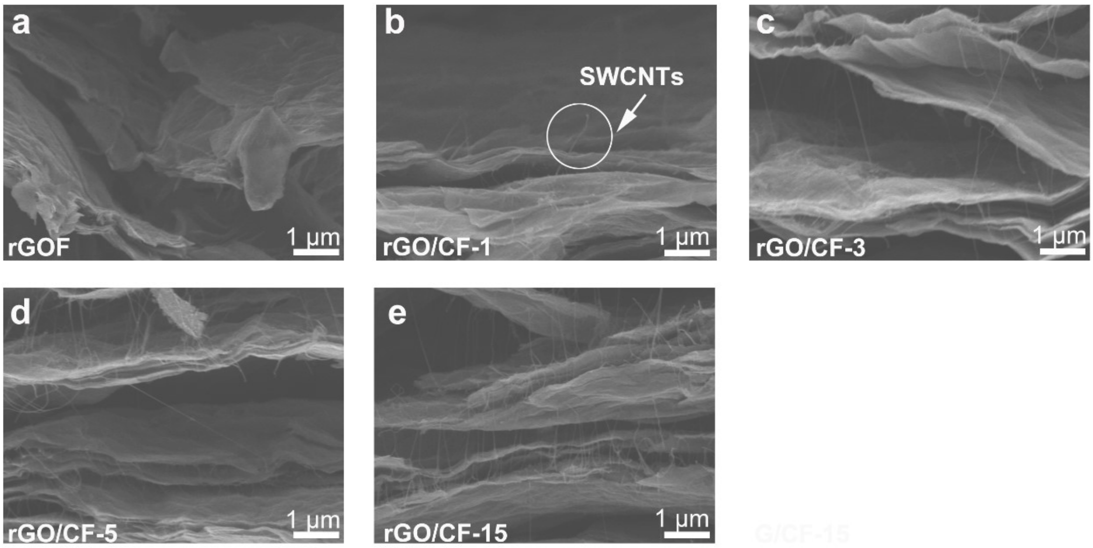

Fig. S1. Higher magnification SEM images of (a) rGOF, (b) rGO/CF-1, (c) rGO/CF-3, (d) rGO/CF-5, and (e) rGO/CF-15.

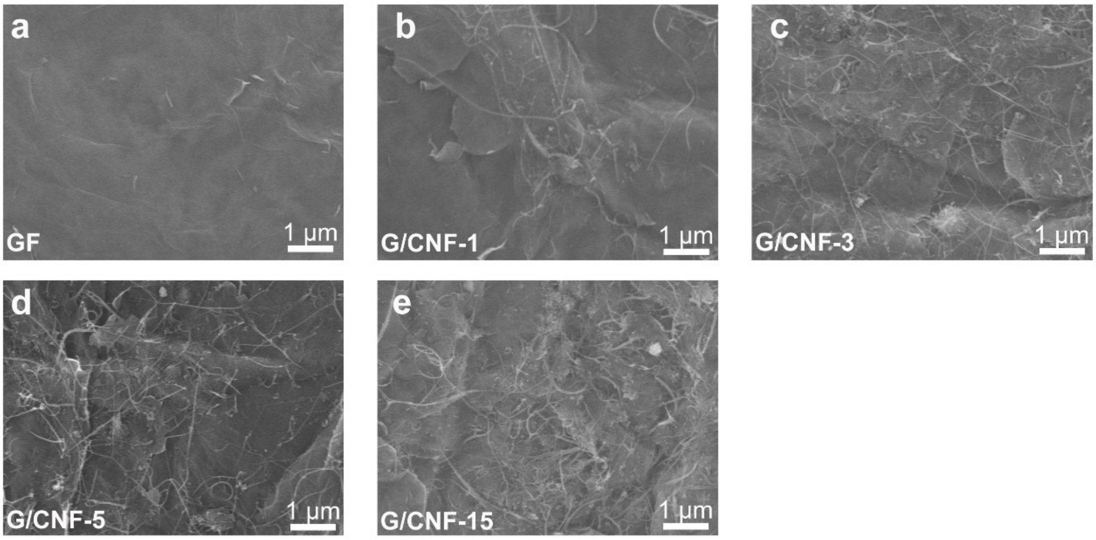

Fig. S2. Higher magnification SEM images for the top view of (a) GF, (b) G/CNF-1, (c) G/CNF-3, (d) G/CNF-5, and (e) G/CNF-15.

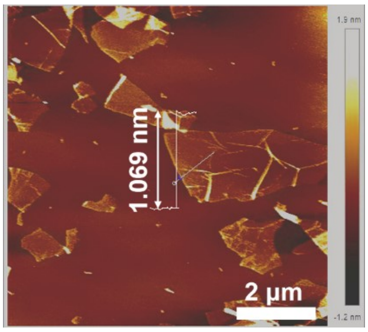

Fig. S3. AFM images of the initial GO sheets

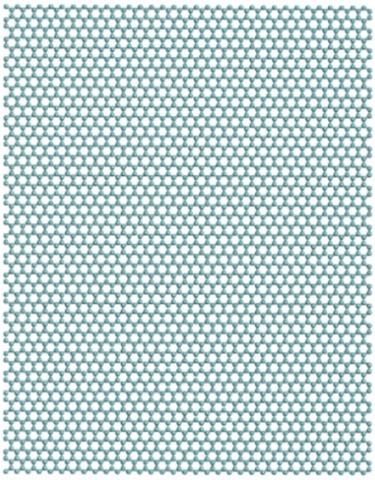

Fig. S4. Models of GF structure (monolayer graphene)

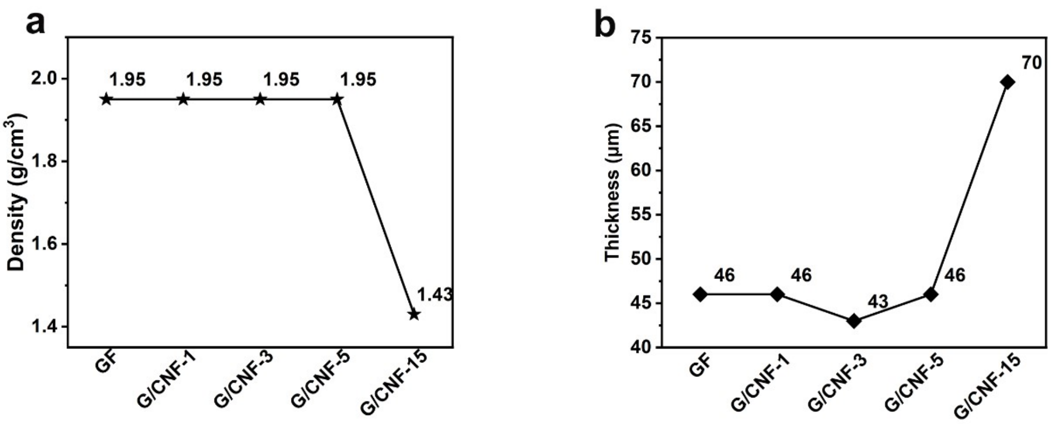

Fig. S5. (a) The density and (b) the thickness of GF, G/CNF-1, G/CNF-3, G/CNF-5, and G/CNF-15.

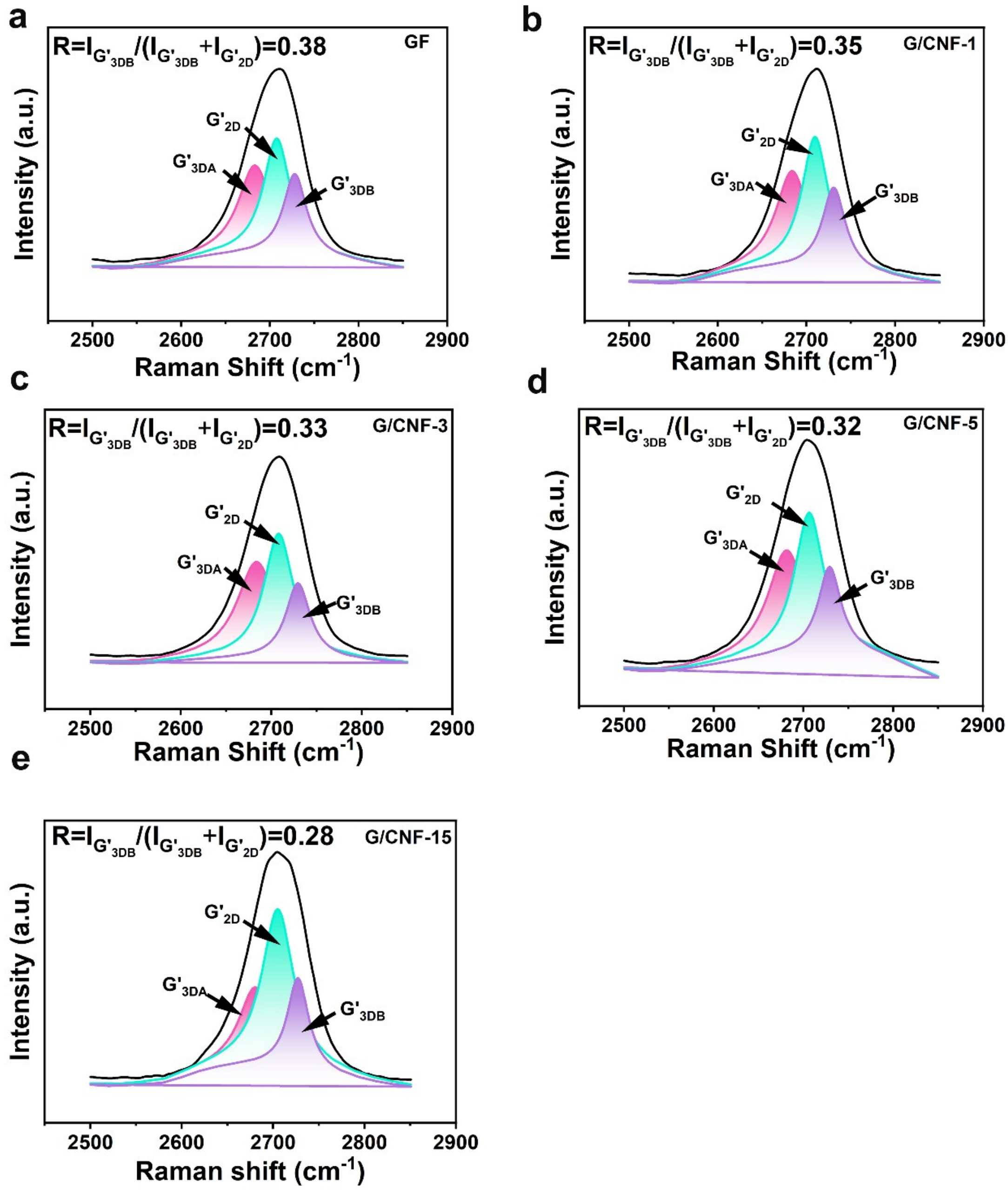

Fig. S6. Lorentzian fitting of the Raman 2D peak of (a) GF, (b) G/CNF-1, (c) G/CNF-3, (d) G/CNF-5, and (e) G/CNF-15.

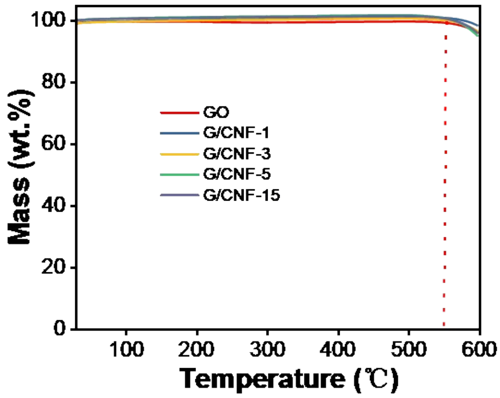

Figure S7. The TGA curves of GF, G/CNF-1, G/CNF-3, G/CNF-5, and G/CNF-15

Table S1. The mass of GO and CNTs contained in the samples

Samples	GF	G/CNF-1	G/CNF-3	G/CNF-5	G/CNF-15
GO (mg)	200	198	194	190	170
CNTs (mg)	0	2	6	10	30

Table S2. The elastic modulus of GF, G/CNF-1, G/CNF-3, G/CNF-5, and G/CNF-15.

Samples	GF	G/CNF-1	G/CNF-3	G/CNF-5	G/CNF-15
Elastic modulus	2.86 GPa	1.41 GPa	247.11 MPa	220.66 MPa	71.69 MPa

Table S3. Bond parameters of CNTs, GF, G/CNF-1, G/CNF-3, G/CNF-5, and G/CNF-15.

Samples	CNTs	GF	G/CNF-1	G/CNF-3	G/CNF-5	G/CNF-15
$c_{sp}{}^{2}$	100835	208448	242791	233100	218783	259731
$c_{sp}{}^{3}$	43092	160632	117111	106388	94920	91659

Calculation formula of the silicon internal standard method:[1]

$2\theta_c = 2\theta_c$ (diffraction angle) + ($2\theta_{si\,(111)}$ (standard)- $2\theta_{si\,(111)}$ (diffraction angle)) S1

$2\theta_c$: standard diffraction angle of samples

$2\theta_c$: diffraction angle of samples in XRD pattern

$2\theta_{si\,(111)}$ (standard): standard diffraction angle of Si, $2\theta_{si(111)}$ (standard)= 28.443°

$2\theta_{si\,(111)}$ (diffraction angle): diffraction angle of Si in XRD pattern

Scherrer equation：

$$L_c = \frac{K\lambda}{\beta_{002}\cos\theta_{002}} \qquad \text{S2}$$

Where K and λ are sample shape constant and X-ray source wavelength (Cu-target, 0.154 nm), respectively. β and θ are full width at half maximum of the (002) peak and Bragg diffractive angle.

Mering-Maire formula:[2]

$$G(\%) = \frac{0.3440 - d_{(002)}}{0.3440 - 0.3354}\times 100 \qquad \text{S3}$$

0.3440: the interlayer spacing of the fully non-graphitized carbon (nm)

0.3354: the interlayer spacing of the ideal graphite crystallite

$d_{(002)}$: the interlayer spacing derived from XRD (nm)

The L_D (nm) can be calculated from Raman I_D/I_G value using:[3]

L_D^2 (nm^2)=(1.8±0.5) $\times 10^{-9}$ $\lambda^4 \times (I_D/I_G)^{-1}$ S4

λ: wavelength (532nm) of laser in Raman testing

n_D (cm^{-2}) can be evaluated using:[4]

n_D (cm^{-2}) =(1.8±0.5) ×10^{22} × λ^{-4} × (I_D/I_G)　　S5

λ: wavelength (532nm) of laser in Raman testing

crystalline domain size (*L*a):[5]

L_a= (2.4 × 10^{-10}) × λ^4× (I_D/I_G)$^{-1}$　　S6

λ: wavelength (532nm) of laser in Raman testing

Raw Materials

Single-walled CNTs (TNST, 5-30 μm length, diameter<2 nm, Purity>95 wt%) were obtained from Chengdu Organic Chemicals Co. Ltd., Chinese Academy of Sciences without further purification. Graphene oxide (GO) was created by modified Hummer's method in our prior reports.

The calculation of thermal conductivity

The thermal conductivity of the film was calculated using the equation (E1):

$$\kappa=\alpha \times C_p \times \rho \quad (E1)$$

where κ, α, C_p, and ρ represent thermal conductivity, thermal diffusivity, specific heat, and sample density. The C_p of all samples is 850 J/ (Kg·K). The laser flash technique (LFA 447 NanoFlash and LFA 467 NanoFlash, Germany) was employed to determine the thermal diffusivity. The density of the film was explored by assessing the mass, thickness and radius of the film

X-ray photoelectron spectroscopy peak fitting process:

Firstly, the test data is imported into the XPS analysis software (XPS peak). Then, we set the background between 282 eV and 297 eV and the background type is set to Shirley. The FWHM of all samples is 0.79 (284.7 eV) and 2.26 (285.4 eV), respectively. The FWHM of all samples is formed by fitting at the binding energy of 290.3 eV. Furthermore, the Lorentzian-Gaussian at different binding energies is set to 20%. The final fitting results of the XPS C1s spectrum are obtained after optimization.

References:

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