Zipping, entanglement, and the elastic modulus of aligned single-walled carbon nanotube films

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Reliably routing heat to and from conversion materials is a daunting challenge for a variety of innovative energy technologies—from thermal solar to automotive waste heat recovery systems—whose efficiencies degrade due to massive thermomechanical stresses at interfaces. This problem may soon be addressed by adhesives based on vertically aligned carbon nanotubes, which promise the revolutionary combination of high through-plane thermal conductivity and vanishing in-plane mechanical stiffness. Here, we report the data for the in-plane modulus of aligned single-walled carbon nanotube films using a microfabricated resonator method. Molecular simulations and electron microscopy identify the nanoscale mechanisms responsible for this property. The zipping and unzipping of adjacent nanotubes and the degree of alignment and entanglement are shown to govern the spatially varying local modulus, thereby providing the route to engineered materials with outstanding combinations of mechanical and thermal properties.

Nanostructured materials provide unique combinations of properties that promise performance breakthroughs for applications ranging from energy conversion to data storage and computation (1–4). In many cases it is the very unusual combination of two properties (2), neither of which is an extreme value when considered alone, that leads to adoption and major performance benefits. An example is the search for a mechanically compliant thermal conductor that can, for example, link semiconductor materials with the metals used for heat spreading and exchange. A particularly compelling case is thermoelectric materials whose performance beneﬁts when considered alone, that leads to adoption and major performance breakthroughs for applications ranging from energy conversion to data storage and computation. Theoretical and experimental studies have suggested that large differences between the modulus of the crust and middle layer are due to their morphological variations. To understand the mechanisms governing the mechanical response of the single-walled CNT films, we use two separate simulation methods. The first is a simple model for cellular solids. Because carbon nanotubes are low-density solids, previous studies have shown that the mechanical response and nanostructure of ﬁlms closely resemble those of a foam or cellular solid (10, 14).

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Vertical aligned carbon nanotube (CNT) films may combine mechanical compliance with high thermal conductivity (7–18), but there have been few reports of the in-plane modulus of these films and little physical explanation for the wide range of the data (2–300 MPa) (19–25). Our previous data for the thickness-dependent in-plane modulus of multiwalled CNT films indicated a strong dependence on the nanotube density and alignment (21), which are linked to the detailed growth details (26–28). Other approaches, such as mesoscopic simulations or atomistic models, found that CNT networks exhibit unique self-organization, including bending and bundling (29–31). Therefore, relating the nanoscale morphological details to the mechanical properties is critical.

Combined experimental, theoretical, and computational techniques applied to the more complex and fundamentally challenging single-walled CNT system are presented in this paper, along with the in-plane data for the modulus for single-walled CNT films. Because single-walled CNT films have higher densities and smaller tube–tube distances, these films exhibit more complex dynamics and tube–tube interactions than multiwalled CNT films. We present coarse-grained molecular simulations, and this

Significance

Aligned carbon nanotube films promise the unusual combination of high thermal conductivity and mechanical compliance. Here, the mechanical compliance of single-walled nanotube films has been measured and linked to their morphology and microscopic motions, including zipping, unzipping, and entanglement. The physical mechanisms governing the mechanical response include binding forces or van der Waals interactions, with the dominant mechanism depending on the nanotube density and alignment. The dependence of film morphology on mechanical modulus explored here provides the foundation for modeling of a variety of other properties including thermal and electrical conductivity.

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been widely used to estimate the modulus of foams (35). Because the film structure consists of angled tube segments rather than perfectly horizontal and vertical beams, we present a modified cellular model in which trusses connect the corners and the center of each unit cell, as shown in Fig. 2A (SI Text) (7). The aspect ratio (AR) of a unit cell is related to the angle made by the trusses with respect to the horizontal plane, where $AR = H/L = \sqrt{2} \tan \theta$. By considering the bending of the trusses in response to horizontal and vertical forces applied to the film, we can predict the in-plane modulus ($E_{1,\text{cell}}$) and out-of-plane modulus ($E_{2,\text{cell}}$) of the film as

$$E_{1,\text{cell}} = C \sin \theta \cos^4 \theta \cdot \left(2 - \cos^2 \theta\right)^{-1}, \quad [1]$$

$$E_{2,\text{cell}} = C \sin^3 \theta, \quad [2]$$

with $C = \alpha E_{\text{ref}} I_{\text{ref}} A_{\text{ref}}^{-2}$, where $E_{\text{ref}}$, $I_{\text{ref}}$, and $A_{\text{ref}}$ are the Young's modulus, moment of inertia, and cross-section area of each truss, respectively, and $\alpha$ is a numerical constant. The volume fraction occupied by the nanotubes is $f_{\text{nan}} = A_{\text{nan}}/HL^2$, where $l$ is the length of the truss. Because nanotubes tend to bundle together, the trusses in our cellular model do not necessarily represent individual nanotubes. Hence, $I_{\text{nan}}$ and $A_{\text{nan}}$ in our model are unknown, and constant $C$ in Eqs. 1 and 2 is treated as a fitting parameter. Fig. 2B plots the predicted dependence of the modulus on the AR at different volume fractions ($f$) of nanotubes. Our model predicts the $f_{\text{nan}}^2$ dependence of the film moduli as the Gibson and Ashby model, but shows a stronger dependence of the film moduli on the AR. The prediction is largely consistent with our experimental data on the film modulus as well as SEM estimates of the AR and $f$. The ratio between the predicted $E_{1,\text{cell}}$ of the middle and crust layer is consistent with the measurements, which is in the range of 30–60. The cellular model provides insight into the overall mechanical behavior of the CNT array. This model assumes that the elastic deformation of the film is entirely caused by truss bending, which is not precisely correct in accounting for the complex morphology in the CNT array. Nonetheless, we apply the cellular model here owing to its simplicity and the possibility, when combined with the coarse-grained approach also developed in this paper, to provide insight into nanotube alignment by means of the AR parameter. In reality, the nanotubes are held together by weak van der Waals forces, and zipping of nanotube bundles may play a role in the elastic compliance of the film. To remove the empirical fitting parameter $C$ and to gain a deeper understanding on the realistic structure and dynamics of the nanotubes in the films, we use a coarse-grained molecular simulation.

In the coarse-grained molecular simulation, each nanotube is modeled as a fiber discretized into a chain of nodes. Neighboring nodes on the same fiber interact to give the bending and stretching stiffness of the nanotube, whereas nodes on different fibers interact through van der Waals forces (31) (SI Text). In this model, the range of AR (1.4–4.2) and $f$ (2–12%) of interest are determined from the characterization results including image processing (7, 9, 11, 13). The moduli calculated using the simulation $E_{1,\text{sim}}$ and $E_{2,\text{sim}}$ are shown in Fig. 2B. Fig. 3 shows that the molecular simulation is qualitatively similar to the SEMs, where the middle layer is more aligned compared with the crust. The average tube orientation $\theta$ in the relaxed structure is determined by comparing with the cellular model. A consistency is observed between the molecular simulations, the cellular model (with a fitting parameter $C = 60,000$), and our experimental measurements, as shown in Fig. 2B. Better agreement is observed particularly at AR < 3.0, where bending is dominant rather than van der Waals forces. However, due to the cancelation effect of the van der Waals interaction and bending energy, the overall trend of the curve (Fig. 2B) remains qualitatively the same. The simulation predicts that the moduli in both directions scale with $f^{1.3–2.0}$, depending on the AR, consistent with the $f^2$ dependence predicted by the cellular model. A better-aligned film with greater AR has not only lower in-plane modulus, which is preferable for the interface applications, but also higher out-of-plane modulus (SI Text). This is because longer beams are weaker in bending, so as the AR increases, the lengthened beams bend more easily in the direction perpendicular to their axis (Fig. 24). If the beams are aligned better, they are more difficult to stretch in the out-of-plane direction.

The molecular simulations also provide more details about the mechanisms of nanotube deformation when the film is subjected to an elastic strain. There are many complex deformation modes not considered in the cellular model. For example, bundles of nanotubes may move together, a bundle formed by several nanotubes may partially zip (Fig. 4A) or unzip, two nanotubes or bundles may rotate around a contact point (Fig. 4B), and nanotubes within a bundle (oriented perpendicular to the straining direction) may slide relative to each other.
To probe the importance of the different deformation mechanisms, we quantify the relative contributions of bending, stretching, and van der Waals energies to the total energy and predict modulus by measuring the average and curvature of these energy contributions as a function of applied strain, respectively. For most films, the van der Waals energy is the dominant contribution to the total energy of the film, meaning that the energy gained in bundling together nanotubes and forming the film structure during the relaxation steps greatly exceeds the energy cost due to nanotube bending and stretching. However, once the film structure has been formed, either bending energy or van der Waals interaction dominates the effective modulus under applied strain, depending on the CNT morphological details (Fig. 4 C and D). For example, for a film of AR = 1.4 and f = 4%, representing the crust layer, the relative contributions of bending, stretching, and van der Waals energies to the total modulus are 72%, 5%, and 23%, respectively. In this case, bending is the dominant contribution in determining the total modulus. However, for a film of AR = 3.5 and f = 2%, representing the middle layer, the contribution of van der Waals energy to the modulus rises up to 60%. Therefore, the contribution of van der Waals interactions gains importance as the nanotubes become more aligned or as the volume fraction increases, because both effects promote the nanotubes to zip into bundles (Fig. 4E).

These results also suggest that the cellular model, which only includes bending forces, is most accurate for structures with low AR and low f (which is the case for many multiwalled CNT samples), although the good agreement between the models suggests that, when fitting parameters are allowed, it is still applicable to higher AR films.

The combination of data and two approaches advances the understanding of the nanostructural effect on the mechanical properties of single-walled CNT films. This understanding is essential for tuning the properties of nanotube films by engineering

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**Fig. 2.** (A) Schematic of a cellular model unit cell comprising eight trusses. The gray region depicts the elastic bending of one truss at an angle θ between the vector along the truss and the plane of the horizontal. (B) Predicted in-plane (E₁) and out-of-plane modulus (E₂) using the cellular models (lines) and coarse-grained molecular simulations (squares) with varied AR and f. The error bars of the simulation results represent variations in the modulus over repeated calculations of the same initial conditions. The line colors indicate the different f (2%, 6%, and 12%). The circled regions include the experimental data of single-walled and multiwalled CNT films (7, 21).

**Fig. 3.** Examples of SEM images (A–C) and simulation snapshots (800 Å × 800 Å × 400 Å) after the relaxation (D–F) of the crust and middle. (A) Cross-sectional SEM images of a single-walled CNT film showing aligned nanotubes in the middle region and (B) randomly oriented nanotubes in the crust region, respectively. (C) Top view of the crust layer. The crust has a value of AR of 4.2 and f of 2% whereas the middle layer has AR of 3.5 and f of 4–8%. In the simulations, the nanotubes are constructed using a specific density and orientation to represent the different film morphologies. (D) Simulated aligned nanotubes (AR = 4.2, f = 2%) and (E) the entangled nanotubes (AR = 2.8, f = 4%). (F) Top view of the entangled nanotubes (AR = 1.4, f = 4%). SEM images of each region of the film are analyzed using an image analysis procedure. The selected SEM images are representative of those obtained for each region.
their nanostructure, which can be achieved by altering the synthesis conditions or by adding surfactant molecules. The development of self-consistent simulations and the experimental data can provide guidance on detailed simulation of other material properties including the effective thermal and electrical conductivities. The approach presented here is also applicable to a wide range of films with a fibrous nanostructure, such as films made of nanowires and microwiskers.

**Methods**

**Coarse-Grained Nanotube Simulation.** We adopt a coarse-grained molecular model in which each nanotube is represented by a set of nodes. The interaction between neighboring nodes on the same chain is designed to reproduce the bending and stretching response of an elastic tube with Young’s modulus $E_{\text{tube}}$, inner radius $r_{\text{in}}$, and outer radius $r_{\text{out}}$. A Lennard-Jones (LJ)-type interaction is introduced to account for the van der Waals attraction and steric repulsion between nanotubes. The potential energy as a function of nodal positions $(r, f)$ can be written as

$$V(r, f) = \sum_{i \neq j} \frac{1}{2} k_b (|r_i - r_j| - b_0)^2 + \sum_{i} \frac{1}{2} k_b \left[ \frac{(r_i + 1 - r_i)}{|r_i + 1 - r_i|} + 1 \right]$$

$$+ \sum_{i} \left( \frac{C_12}{|r_i - f_i|} - \frac{C_6}{|r_i - f_i|^6} \right)$$

[3]

The first summation is over all neighboring nodal pairs on the same nanotube; the second summation is in which $i$ is the neighbor of nodes $i-1$ and $i+1$ on the same nanotube; the third summation is over all nodal pairs that do not belong to the same nanotube (where $i \neq |i| \geq 1$). $k_b = E_{\text{tube}} A_{\text{tube}} l_0$, and $k_b = E_{\text{tube}} A_{\text{tube}} l_0$, where $A_{\text{tube}} = \pi r_0^2$ is the moment of inertia of the tube cross-section, and $l_0$ is the discretization length of the nanotube. Here we adopt $r_{\text{out}} = 5 \text{ Å}$, $r_{\text{in}} = 4.4 \text{ Å}$ by using the effective tube thickness (37). The LJ coefficients between the nodes correspond to the LJ coefficients describing the interaction between individual carbon atoms. Specifically, $C_{12} = N_1 C_{12}$ and $C_6 = N_6 C_6$, where $N_i$ is the number of carbon atoms represented by each node (36), $C_{12} = 2.516 \, \text{eV} \cdot \text{Å}^{-1}$, and $C_6 = 1,228.8 \, \text{eV} \cdot \text{Å}^{-6}$ (38). For single-walled CNTs, we estimate $N_i$ to be $2 \pi r_{\text{out}} l_0 A_{\text{c}}$, where $A_{\text{c}}$ is the average area covered by each carbon atom. The nanotubes are initialized as straight lines randomly positioned in the simulation cell. The number of nanotubes and the length of each nanotube are selected to achieve the desired density. The initial nanotube orientations are parallel to the $x$, $y$, and $z$ axes, respectively, relaxing the structure again, and computing the stress change. The maximum strain applied to the simulation cell is $\pm 0.1\%$, which is also the maximum applied strain in our experiments. The effective modulus of $E_{1,\text{sim}}$ and $E_{2,\text{sim}}$ is computed from the components of the elastic stiffness tensor. For example, $E_{1,\text{sim}}$ corresponding to the equivalent value of $E_{1,\text{exp}}$ in the current work is

$$E_{1,\text{sim}} = \frac{C_{111}}{C_{111} - (C_{111} C_{12} - C_{12} C_{11}) (C_{11} C_{22} - C_{12}^2)^{-1} C_{12}}$$

$$- \frac{(C_{22} - C_{12} C_{22}) (C_{22} - C_{12} C_{22})^{-1} C_{22}}{C_{22}}$$

[4]

where $\sigma_1$ and $\sigma_2$ are assumed to be zero because of the free surfaces of the single-walled CNT films. Because vertically aligned CNT films have entangled morphologies showing foam-like behavior (10, 15, 17, 18), we assume that the film has a Poisson’s ratio of zero. Thus, the $E_{2,\text{sim}}$ is assumed to be $C_{22}$. The model accounts for the nanotube morphology by incorporating various density and alignment parameters (i.e., $\Delta$, $A$, and $\phi$). The coarse-grained molecular simulation predicts the mechanical response of single-walled CNT films depending on the film nanostructures and the intrinsic mechanical behavior of nanotubes.

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**Supporting Information**

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**SI Text**

**The Growth Procedure of Vertically Aligned Single-Walled Carbon Nanotube Films.** Vertically aligned single-walled carbon nanotube (VA-SWNT) films have been successfully grown on fabricated cantilevers using an alcohol catalytic chemical vapor deposition system, which has the advantages of high quality and selectivity (1). To enhance the yield of VA-SWNT films, a 50-nm SiO2 layer is first grown on the substrates. The wafer is cleaved into many pieces to allow each piece to be processed individually to achieve different thicknesses of VA-SWNT films. A thermal evaporator deposits the catalyst films of 10-nm-thick Al and 0.2-nm-thick Co. VA-SWNT films are then synthesized using ethanol (1.3 kPa) as the carbon source at 800 °C for different time frames, such as 1, 3, and 10 min. During the synthesis, CNT film thicknesses are controlled by optical absorbance, using the relative transmitted intensity of the incident laser. Fig. S1L shows a typical Raman spectrum of the material grown on the cantilevers (red lines). A strong radial breathing mode (RBM) ranging from 100 to 300 cm\(^{-1}\) confirms the existence of SWCNTs; the weak disorder mode at 1,340 cm\(^{-1}\) suggests a high degree of crystal perfection in the produced SWCNT. A transmission electron microscopy (TEM) image of SWCNTs further reveals that the growth provides almost 100% SWCNTs with an average diameter about 2 nm, as shown in Fig. S1B. Based on this value and the mass density of the VA-SWNT film, the number density of VA-SWNTs is calculated to be around 10\(^{12}\) cm\(^{-2}\) (2). Thus, the average tube–tube spacing is ~10 nm. Because SWCNTs often form bundles, the real bundle–bundle distance could be a few tens of nanometers.

**Measurement Method.** The resonant frequency of a single-material, fixed-free beam for given dimensions can be calculated by solving the one-dimensional Euler–Bernoulli differential equation with assumptions including small deflections, linear elastic behavior, and isotropic, homogeneous beams, and constant cross-section. The modulus can be extracted by modeling the Si-CNT cantilevers as two-layer composites, using their resonant frequency. The ratio of the resonant frequency shift to the resonant frequency of the silicon-only beam \(\Delta\) is given by

\[
\Delta = \left( \frac{(EI)_{Si} + (EI)_{CNT}}{(\rho A)_{Si} + (\rho A)_{CNT}} \frac{(\rho A)_{Si}}{(EI)_{Si,0}} \right)^{0.5} - 1, \tag{S1}
\]

where \(E, I, \rho, \) and \(A\) are, respectively, the modulus, the second moment of area, density, and the cross-sectional area of each layer, and \(\Delta\) is the measured ratio of the resonant frequency shift to the resonant frequency of the silicon-only beam. The subscripts \(Si, 0,\) and \(CNT\) denote the silicon-only beam, the silicon layer, and the VA-CNT layer of a Si-CNT composite beam, respectively. Details and validation of this measurement are explained in ref. 3.

Because the crust remains a constant thickness over growth time, a three-layer analysis captures the impact of increasing the thickness of the middle layer in a composite beam using Eq. S2.

\[
\Delta = \left( \frac{(EI)_{Si} + (EI)_{crust} + (EI)_{middle}}{(\rho A)_{Si} + (\rho A)_{crust} + (\rho A)_{middle}} \frac{(\rho A)_{Si}}{(EI)_{Si,0}} \right)^{0.5} - 1, \tag{S2}
\]

where the subscripts \(crust\) and \(middle\) denote the crust and middle layer, respectively. The thickness of the crust layer is determined using SEM images of film cross-sections.

**Cellular Model.** We present a cellular model in which the trusses connect the corners and the center of each unit cell, as illustrated in Fig. S2. The aspect ratio (AR) of a unit cell is related to the angle \(\theta\) the trusses make with the horizontal plane through \(AR = H/L = \sqrt{2} \tan \theta\). The truss has length \(l\) and is oriented at an angle \(\phi\) to the vertical plane of the cell, where \(\cos^2 \phi = 1 - 0.5 \cos^2 \theta\).

The in-plane and out-of-plane modulus can be predicted by considering the behavior of a single truss in a unit cell with height \(H = l \sin \theta\) and width and length \(L = l \cos \theta/\sqrt{2}\). Because this simple model does not include interactions between nanotubes such as zipping or sliding motions, the nanotube behavior remains linearly elastic under the action of a horizontal load \(P = \sigma_L L\) or vertical load \(W = \sigma_L L^2\). Under a horizontal load \(P\), the deflection in the horizontal direction is defined as \(\Delta_2 = P L^3 \cos \phi/12EI_{tub}\), using elementary beam bending theory, and the strain is \(\varepsilon_L = \varepsilon_L \cos \phi L^{-1}\). Thus, the in-plane modulus \(E_{L,cell} = \sigma_L/\varepsilon_L\) calculated as \(E_L = C \sin \theta \cos \theta (2 - \cos^2 \theta)^{-1}\), where \(\sigma_L = A_{tub}/(HL) = 2A_{tub}/\sin \theta \cos \theta\). Under a vertical load \(W\), the deflection in the vertical direction \(\Delta_2 = W L^4 \cos \phi/12EI_{tub}\), and the out-of-plane modulus \(E_{I,cell}^2\) is defined as \(C \sin \theta \).

**Coarse-Grained Molecular Simulation Method.** This paper presents a nanotube simulation, which predicts the mechanical response depending on the film morphology and the intrinsic mechanical behavior of nanotubes. Instead of simulating individual atoms, we adopt a coarse-grained model in which each nanotube is represented by a set of nodes, as illustrated in Fig. S3, where an SWCNT consists of many elastic short nanotubes which act as Euler–Bernoulli beams. The simulation treats bonds between nodes as springs to calculate the energy of each section and uses nodes to describe the locations of unit tubes. The behavior of each section includes bending, stretching, and interactive forces. The model accounts for the nanotube morphology by incorporating density and alignment parameters in the model.

**Coarse-Grained Molecular Simulation Results.** By varying the orientation preference and number density (CNTs/Å\(^2\)), we can define AR and \(f\) of nanotube films to compute the elastic moduli of SWCNT films, as shown in Fig. S4. The resulting in-plane \((E_{I,cell})\) and out-of-plane \((E_{II,cell})\) moduli for various AR and \(f\) range from 1 to 100 MPa and from 5 to 700 MPa, respectively. A denser film displays higher modulus in both directions. A film with greater AR leads to lower \(E_{I,cell}\) and higher \(E_{II,cell}\). This work indicates that \(E_{I,cell}\) and \(E_{II,cell}\) increase as \(f\) \(\rightarrow\) 2\% for individual sets of various AR as a function of \(f\). The results imply that the computed modulus from the coarse-grained simulations is strongly related to the nanostructural characteristics.

**Contributions of van der Waals, Bending, and Stretching Energy to the Modulus.** The curvature of van der Waals, bending, and stretching energy curves over varying strain enables us to separate and quantify the contributions to the modulus. Once the film structure has been formed, either bending energy or van der Waals interaction dominates the effective modulus during applied strain, depending on the CNT morphological details. An example of the energy change curves from the original structure is shown in Fig. S5 for a film with a relatively low volume fraction (AR = 1.4, \(f = 2\%\)). Each component of bending, stretching, and van der Waals energy to the total modulus, respectively, yields 5.40, 0.10, and 0.55 MPa, giving a total modulus of 6.05 MPa. The contributions of each component are about 89%, 2%, and
9%, respectively. Here, the bending energy is a major contribution to the total modulus. Fig. 4 shows how the relative contributions of bending, stretching, and van der Waals change depending on the film structure. For entangled films with lower AR, the van der Waals interactions clearly increase as $f$ increases, because increasing $f$ corresponds to more nanotube interactions (bundling and crossing). The contribution of van der Waals interactions in films with higher AR shows a smaller dependence on $f$. This may be because when tubes are well aligned; the relative interactions are not strongly dependent on $f$. Also, there is much more variation in the resulting structure for the same initial condition. The contribution of van der Waals interactions becomes more important as the nanotubes become more aligned or as $f$ increases.


**Fig. S1.** Characterizations of VA-SWCNT films. (A) Raman spectrum of the VA-SWCNT films measured by 488-nm laser light. (Inset) RBM peaks are shown. The dominant peak at 175 cm$^{-1}$ is found to split into sharp and separate peaks. (B) TEM image of SWCNTs.

**Fig. S2.** Schematic depicting the elastic bending of a truss segment at angles $\theta$ and $\phi$ between the vector along the nanotube and the plane of the horizontal sheet and vertical sheet, respectively.
Fig. S3. Illustrations depicting an individual CNT and snapshots of the coarse-grained molecular simulation. (A) Each nanotube is represented by a set of nodes connected by line segments. (B) The interaction between neighboring nodes is designed to reproduce the elastic behavior of a nanotube with inner radius $r_{in}$ and outer radius $r_{out}$. In reality, the nanotube consists of carbon atoms in a periodic hexagonal arrangement. The number of carbon atoms in a segment tube is used to calculate the van der Waals interaction per a segment. (C) Snapshots of the simulation showing the initial structure (Upper) and relaxed structure (Lower). The example cell size is $800 \, \text{Å} \times 800 \, \text{Å} \times 800 \, \text{Å}$.

Fig. S4. Coarse-grained simulation results of in-plane ($E_{1,sim}$) and out-of-plane ($E_{2,sim}$) modulus of SWCNT films: (A) $E_{1,sim}$ increases with $f$ for various AR; (B) $E_{1,sim}$ decreases with AR for various $f$; (C) $E_{2,sim}$ increases with $f$ for various AR; (D) $E_{2,sim}$ increases with AR for various $f$. The error bars correspond to the SD of the data points after repeating the simulation six times.
Fig. S5. Energy changes from the original structure (at strain = 0%). When the original cell is stretched by various strains ranging from -0.1% to +0.1% and then relaxed, the contribution of each component to the total energy changes. Stretching ($E_s$), bending ($E_b$), and van der Waals ($E_{vdw}$) components are represented by gray, red, and black lines, respectively.