Electronic Supplementary Information

Bundling dynamics regulates the active mechanics and transport in carbon nanotube networks

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Methods

Synthesis of vertically aligned SWCNTs. A multilayered substrate ($Al/SiO_2/Si$) is employed for the synthesis of the millimeter long SWCNTs. A 20 nm thick Al buffer layer is first deposited on a SiO_2(100 nm)/Si wafer. The surface is coated with a 0.7 nm thick Co catalyst layer using an e-beam evaporator and then placed inside a quartz tube. The tube is evacuated to 15mTorr and then exposed to a 5% H_2- 95% Ar mixture carrier gas at a pressure of 700 Torr and temperature 850◦C. After the stabilization, high-purity anhydrous ethanol (99.95%) vapor is supplied from the bubbler as a carbon source which results in the growth of vertically-aligned SWCNTs.

Assembly of the sandwiched hybrid superstructure, S/CNN/S. A two-step polymer casting method is use to synthesize the S/CNN/S superstructure. Each step is illustrated schematically in SI (Fig. S1) A bilayer metal contact pad (Ti (5 nm)/Au (150 nm)) is first deposited on the top-side of the as-grown vertically aligned SWCNTs using a sputter coater (Fig. S1a). A spin coater is used to coat the SiO_2 wafer with liquid phase PDMS (weight ratio of 10:1) and then 80% cured. The top-side of the CNN with the metal pad is placed on the surface of the PDMS coating and then fully cured at 110◦C for 90 seconds (Fig. S1b). The SiO_2 wafers are detached from bottom (Fig. S1c and S1d) and the bilayer metal pad is deposited on the bottom-side of the CNN (Fig. S1e). It is finally capped with the second PDMS layer by pouring liquid PDMS and then curing at 110◦C for two hours (Fig. S1f-h).

Electrical and Thermal Chracterization. A Janis ST-500 electric probe station connected to a Keithley 240 sourcemeter was employed for electrical characterization. The thermal conductivities of the three different SWCNTs networks were measured by using standard laser

flash thermal constant analyzer (UlVAC TC7000- HNC).

Coarse-grained computations. The radius dependent stiffness for SWCNTs are based on prior atomic simulation [36]. All stiffness used in our computations are for (8, 8) SWCNTs (stretching k_s = 626.46 eV/Å, bending k_b = 9063.02 eV/Å and torsional k_t = 6252 eV/ Å). The spring length connecting two neighbor beads has uniform length Δl = 2 nm and is small enough such that the orientation dependence of the interaction between beads can be ignored. The cohesive energy (−0.845 eV/nm) and the equilibrium inter-tube distance (1.4 nm) between parallel (8, 8) SWCNTs are readily available from the universal graphitic potential and serve as the two parameters for fitting the coarse-grained Lennard-Jones pair potential, σ = 1.534 nm and ε = 0.81 eV [34, 35]. The CNN is generated as described in the text and is stretched in a strain-controlled fashion by uniformly displacing the fixed ends of the SWCNTs (Z-direction). The Poisson's ratio in the experiments is negligible to simulate this, the cross-section area (X − Y plane) of the periodic computational cell is held constant. Densification is simulated by laterally compressing the computational cell while maintaining the distance between the fixed ends of the SWCNTs. The time step for the dynamics is fixed, Δt = 5 fs. To capture the changes in the network topology during physical loading, we track the bundle the maximum bundle size by sectioning the network into 1000 cross sections. Within each section, two SWCNTs are deemed to belong to one bundle if the inter-tube distance is smaller than the cutoff distance between the average first and second neighbors in an idealized hexagonally packed SWCNT bundle.

Figure S1.

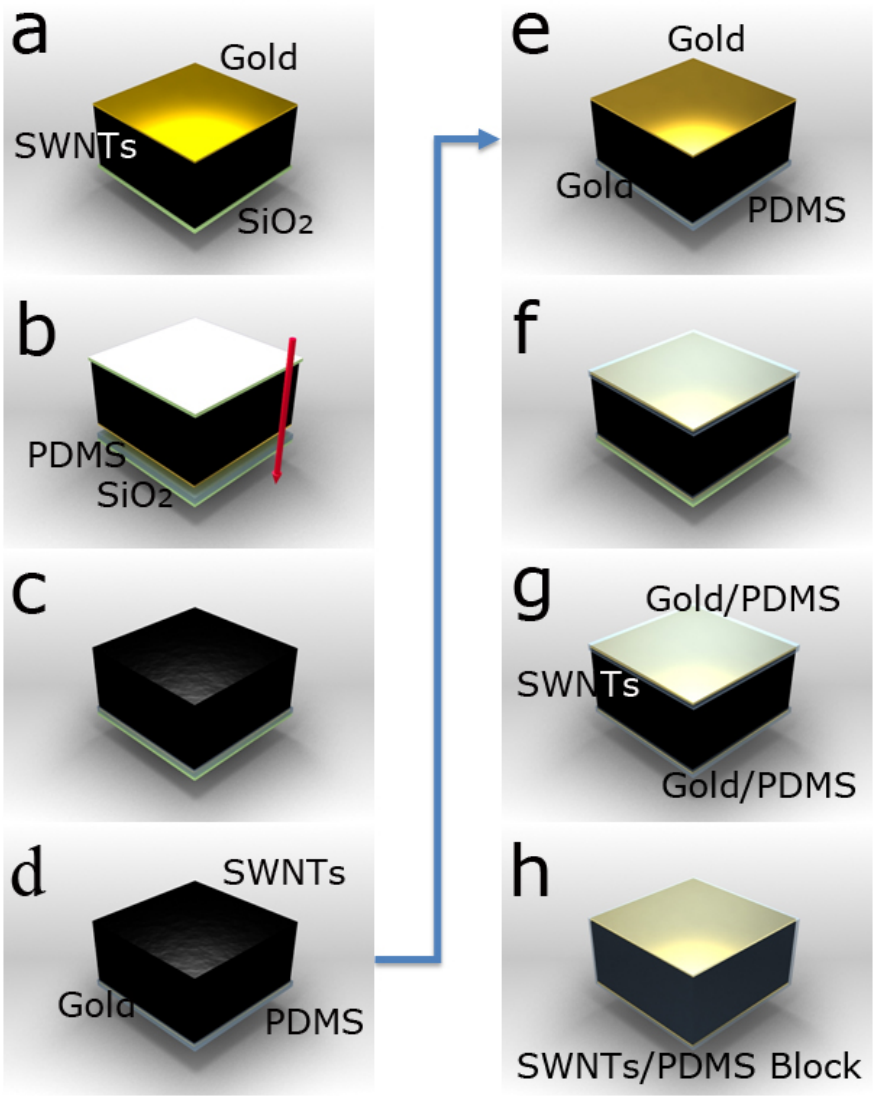

Figure S1. Schematic drawing of fabrication procedure for PDMS/vertically aligned SWNTs/PDMS/ hybrid structure. (a) Deposition of Ti (5 nm)/Au (150 nm) on as – grown vertically aligned SWNTs. (b) Putting the top side of Ti/Au/SWNTs on 80% cured PDMS thin film (300 μm thick). (c) After complete curing of PDMS thin film, removing the SiO_2 wafer for SWNTs growth. (d) Detaching the SiO_2 wafer for PDMS thin film coating. (e) Second deposition of Ti/Au contact pad on top side of first transferred SWNTs/PDMS structure. (f) Putting the top side of Ti/Au/SWNTs/PDMS structure on 80% cured PDMS thin film. (g) After 100% curing of PDMS thin film, removing the SiO_2 wafer for PDMS thin film coating. (f) Infiltrating PDMS and curing.

Figure S2.

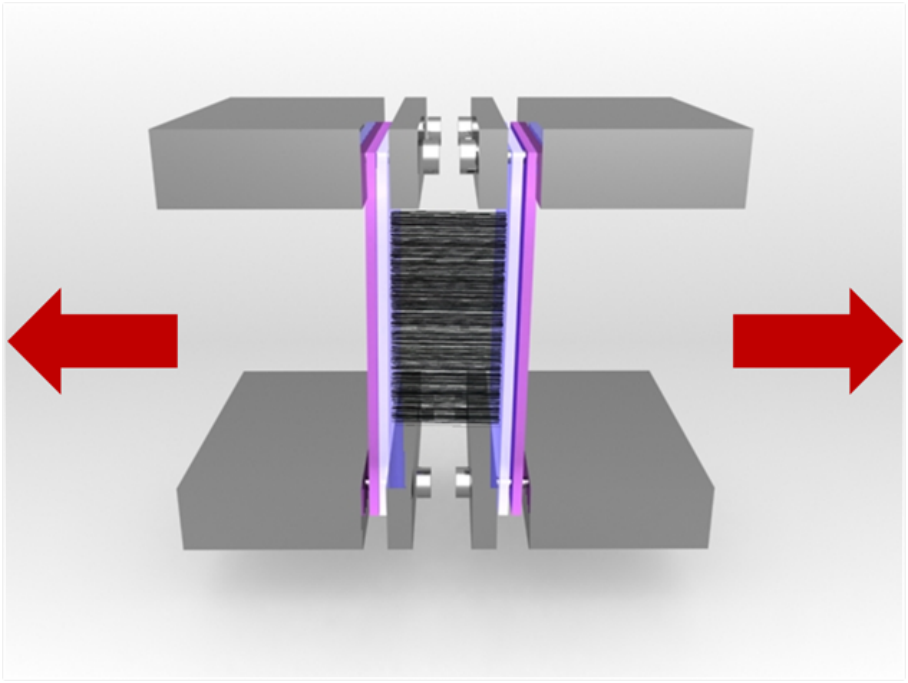

Figure S2. Schematic drawing of stretching of vertically aligned SWNTs networks.

To clarify the applied strain for densified CNN, the relative strain of CNN was investigated by Raman spectroscopy. First, the strain of stretched CNN was calculated by the amount of stretching and the relative strain was investigated by comparison of the peak positions of G band spectra stretched and as-synthesized CNN). Then, the relative strain of densified CNN was measured by comparison of the peak positions of G band spectra (densified and as-synthsized CNN).

Figure S3.

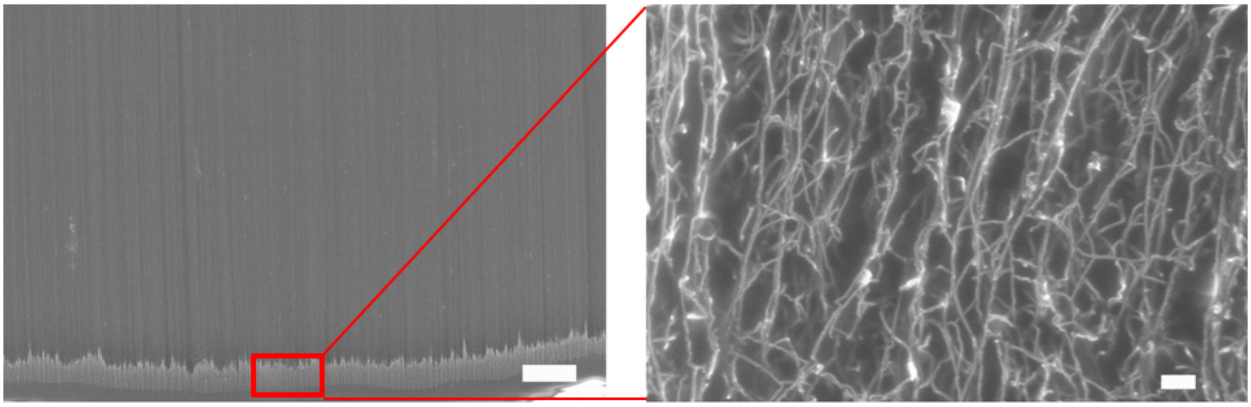

Figure S3. Cross – sectional SEM images of interface of PDMS and SWNTs. Left image shows a low magnification SEM image of bottom side of PDMS/SWNTs/PDMS sandwich structure. The scale bar is 200 μm. The image shows the interface between PDMS and SWNTs. Right image shows a high magnification SEM image of SWNTs anchored into PDMS matrix. The scale bar is 200 nm.