Supplementary Information

Aligned Carbon Nanotube, Graphene, and Graphite Oxide Thin Films via Substrate-Directed Rapid Interfacial Deposition

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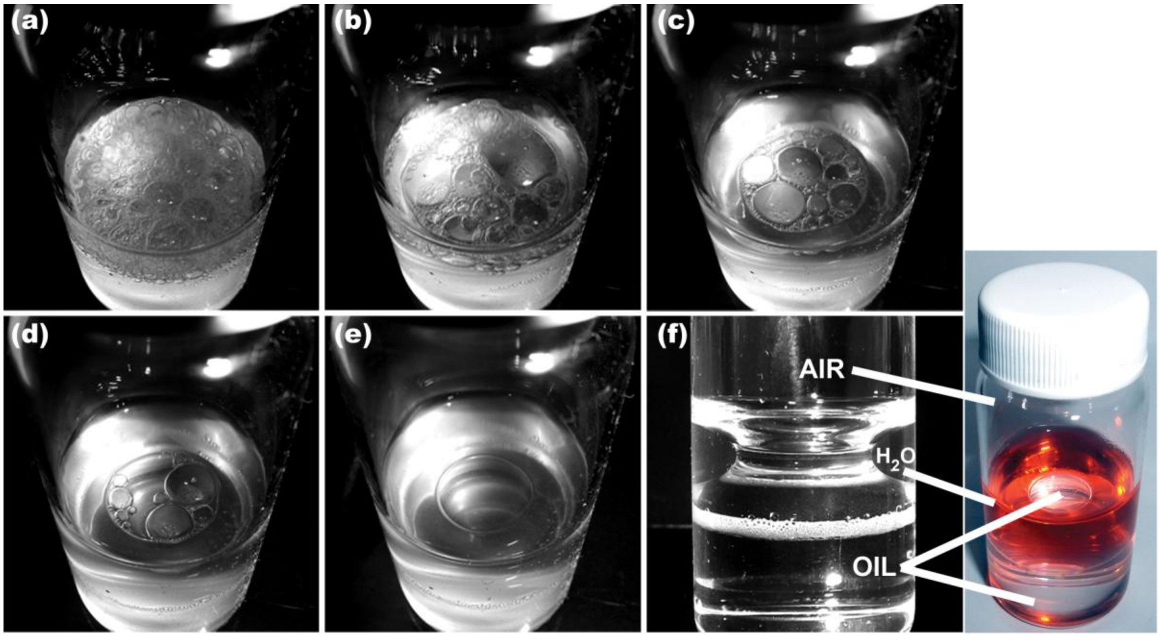

Fig. S1 Photographic sequence shows the coalescence of droplets and the formation of a catenoid. A thin liquid film spreads up a hydrophilic wall when water and an oil such as a dense halogenated hydrocarbon are vigorously agitated in a glass vial and left standing. Optical photographs show: (a) the presence of the initial unstable emulsion of droplets; (b) coalescence of small droplets leading to larger droplets; (c) water ring pressed against the glass wall; (d) droplets coalescing inside the water ring; and (e) when coalescence stops the water ring remains. (f) The profile of the water ring shows that it is actually a catenoid – the water layer adopts this geometrical shape in order to minimize its surface energy. A water soluble red dye shows that the inner catenoidal channel is comprised of oil.

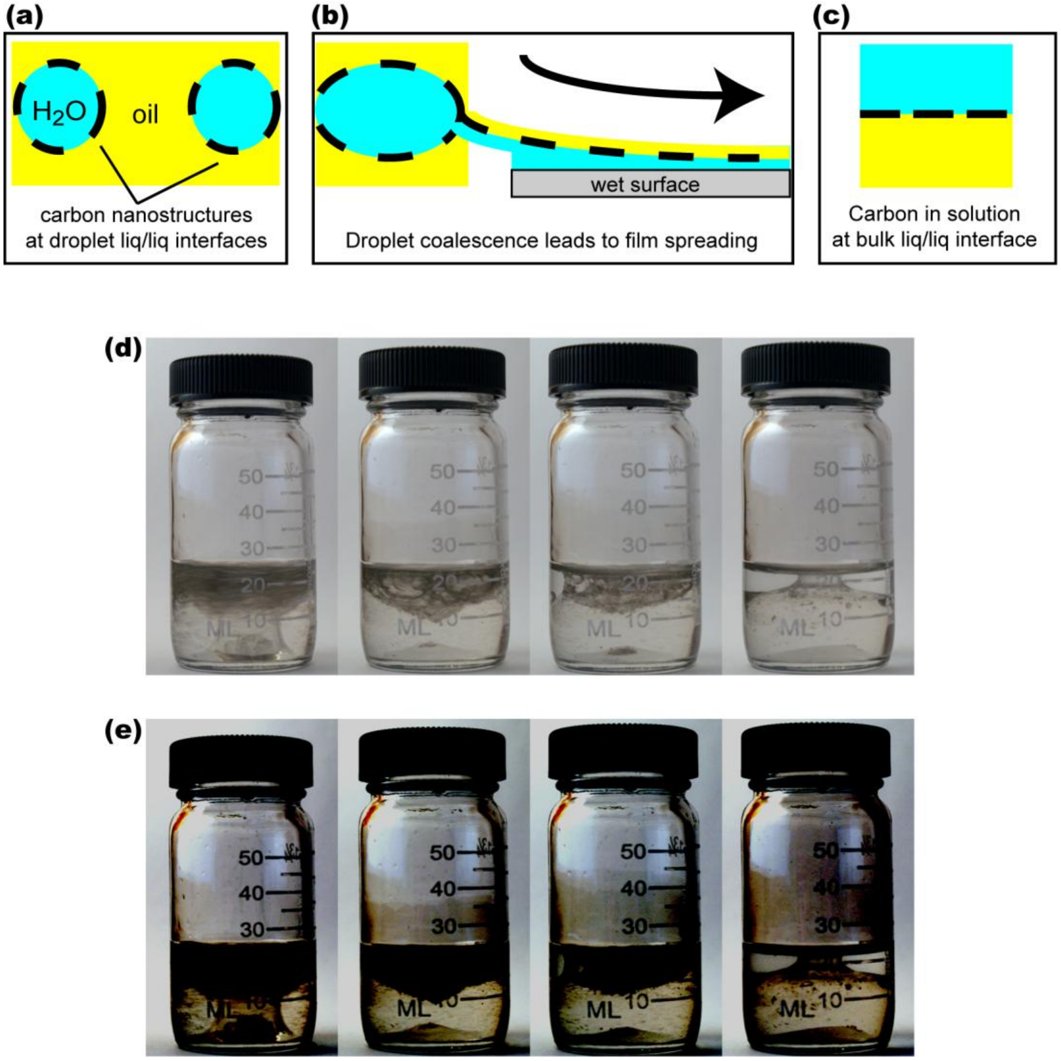

Fig. S2 Schematic diagram depicting the growth mechanism and a photographic sequence that shows the spreading of a SWCNT film. When a Pickering emulsion of carbon nanostructures coalesces it promotes fluid flow. (a) A water in oil emulsion of droplets is partially stabilized by carbon nanostructures. (b) Upon standing, the droplets coalesce and expel excess oil and nanostructures (left), and this generates an interfacial surface tension gradient that leads to film deposition over a wet substrate (right). (c) A reservoir of solid materials remains between the bulk liquid phases after film growth stops, containing excess sheets that can be used to coat additional substrates. (d) A transparent film of SWCNTs grows on glass when a Pickering emulsion coalesces; this sequence shows the phase separation of the emulsion and film growth. (e) A digitally retouched sequence aids in visualization.

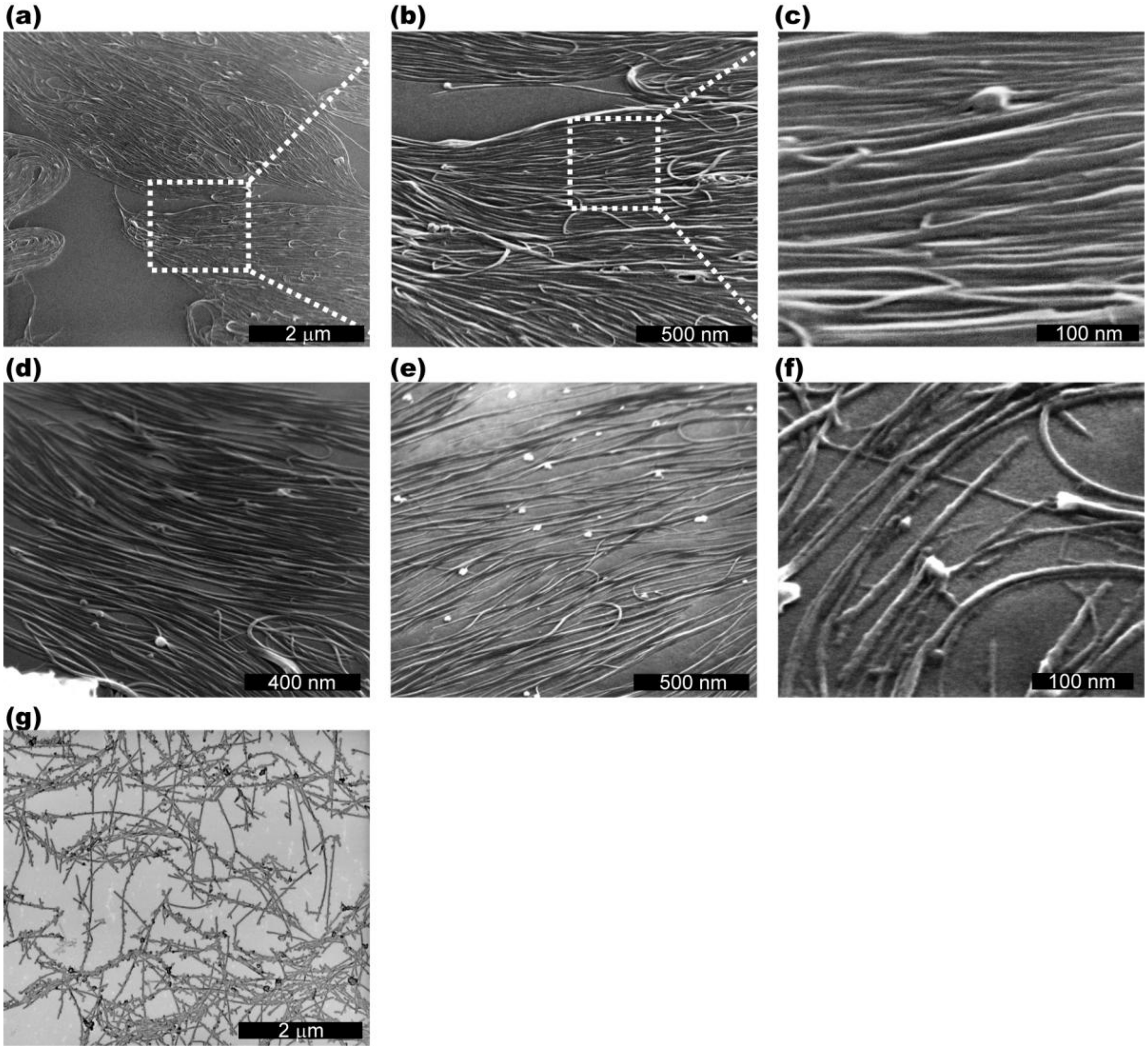

Fig. S3 Tilted scanning electron micrographs show the morphology of a SWCNT film on SiO_2. (a) The film morphology is comprised of SWCNT domains that stack in 2D. (b) Aligned nanotube ropes assemble into a dense array possessing a high-packing density. (c) Magnified view of aligned SWCNT ropes. A less entangled thin film morphology can be obtained by (d) using a 0.025 mg/mL dilute aqueous dispersion of SWCNTs, (d) annealing at 400 C for 6 hr and by (e) utilizing 5 centrifugation cycles during purification. (f) Combining multiple centrifugation cycles and extensive sonication also leads to a low degree of entanglement however it results in a thin film morphology without alignment.

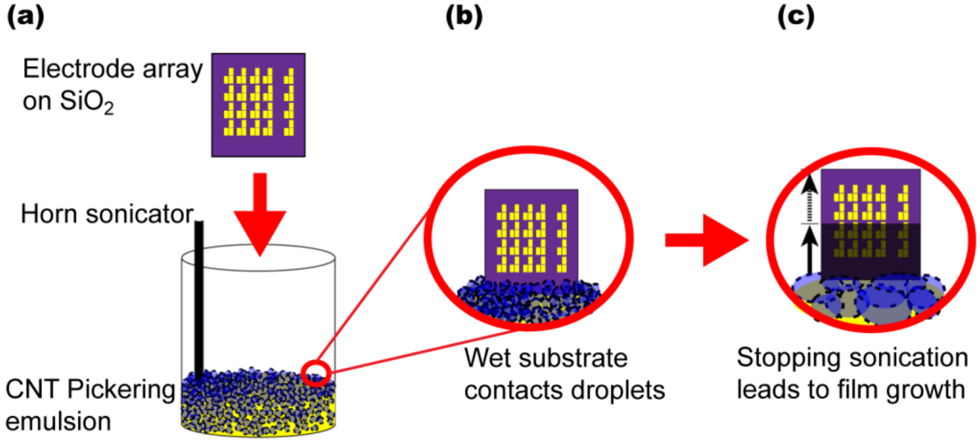

Fig. S4 Process diagram for automated deposition of a SWCNT thin film. (a) An emulsion of droplets stabilized by SWCNTs is produced using a horn sonicator. (b) A wet SiO_2 substrate contacts the emulsion surface. (c) The sonicating tip is turned off and coalescence ensues leading to film deposition.

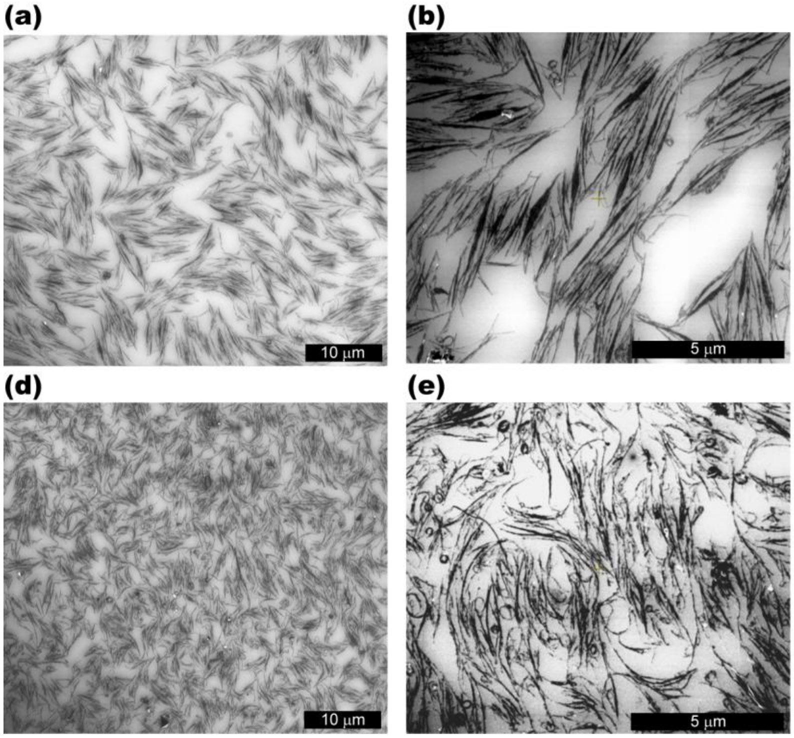

Fig. S5 Scanning electron micrographs of SWCNT films deposited on SiO_2. Film deposition is carried out using a binary immiscible mixture comprised of an aqueous phase containing hexafluoroisopropanol and a halogenated hydrocarbon. This mixture is sonicated extensively prior to film deposition and leads to (a-b) a thin film morphology of well separated ropes and individual carbon nanotubes when using chlorobenzene as the other phase. (c-d) When chloroform serves as the other phase, it leads to a film morphology containing more individual carbon nanotubes.

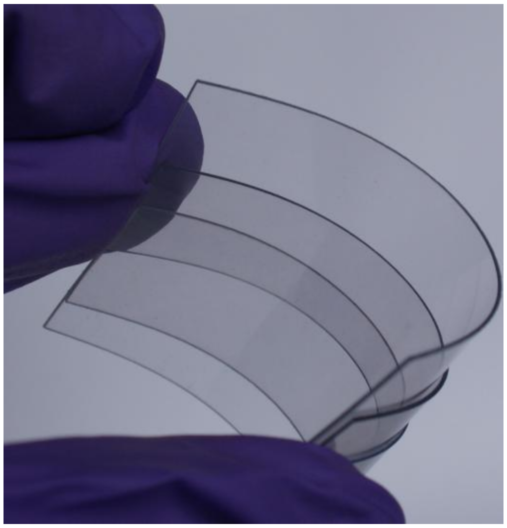

Fig. S6 SWCNT films on flexible plastic slides.

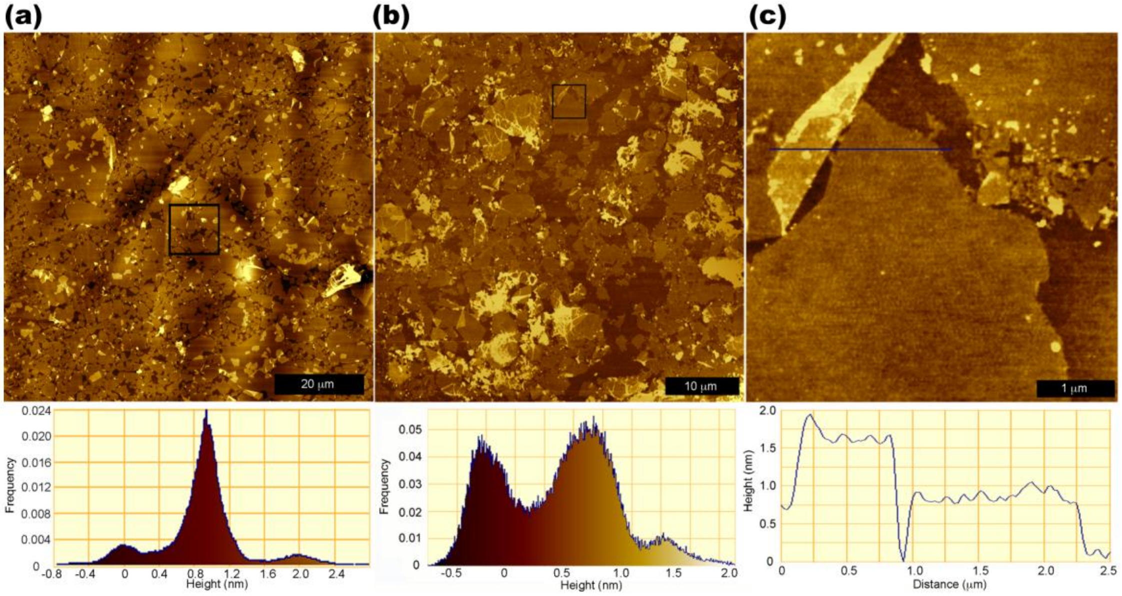

Fig. S7 Representative AFM images, histograms and cross-sectional analyses. (a) The histogram of a GO coating demonstrates a film structure largely comprised of single sheets. (b) CCG film on SiO_2 deposited using anhydrous hydrazine and chlorobenzene leads to a densely packed coating of single sheets. (c) Height profile of a CCG film shows a sheet thickness of 0.75 nm.

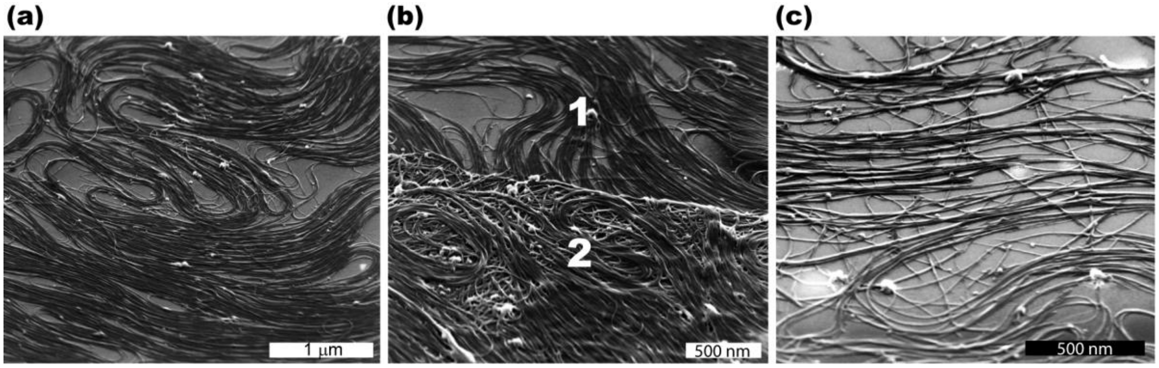

Fig. S8 Tilted scanning electron micrographs of free-standing films of SWCNT collected on SiO_2. (a) A single layer retains alignment after delamination and transfer. (b) The junction between single and double layers. (c) Using an aqueous dispersion ranging in concentration between 0.06 mg/mL and 0.08 mg/mL leads to a less entangled SWCNT free-standing thin film morphology.