Supporting Materials

Solution-Processable Semiconducting Thin-Film Transistors Using Single-Walled Carbon Nanotubes Chemically Modified by Organic Radical Initiators

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Experimental Section:

Radical Reaction: The purified SWeNT@SG 65 tubes were purchased from SouthWest Nanotechnologies (USA) and directly used. To obtain SWNT suspension, 0.3 mg of SWNTs was dispersed in 30 mL of DMF solution via probe-ultrasonication for 30 min (Sonics & Materials Inc., Model: VCX 130). Then, 1 mL of DMF solution containing 25 mg/mL ACN was added to 10 mL SWNT suspension, followed by 30 min ultrasonication. After reacton with ACN, the suspension was filtered through a 0.25 µm PTFE membrane, followed by repeated washing with DMF and acetone to remove the residuals. The powders collected from PTFE membrane were re-dispersed in a 2 wt % of co-surfactants which consists of sodium dodecyl sulfate (SDS) and sodium cholate hydrate (SC) (weight ratio=1:4). The nanotube bundles in the suspension were removed by centrifugation at 20000 rpm for 90 min. The resulting supernatant was then used for fabrication of thin-film field-effect transistors (FETs).

Fabrication of FET devices: The SWNT FETs (SNFETs) were fabricated by drop-casting the suspension of modified SWNTs across two Au electrodes (100 nm thick) pre-patterned on top of SiO2/Si substrate to form conducting channel ~50 µm long and ~25 µm wide. The gate dielectrics SiO2
is 300 nm thick. For the drop-cast procedure, 25 µL of SWNT suspension was dropped onto the devices, followed by drying at room temperature and rinsing of de-ionized water. The procedure was repeated until the density of ACN-functionalized SWNTs is high enough to reach the desired current level.

**Estimation of device mobility:** The effective field-effect mobility is estimated by

\[ \mu = 10^4 \times \frac{I_d}{V_{gs}} \times \frac{L}{W} \times \frac{1}{C_{ox} V_{ds}} \]

Here, \( C_{ox} \) is determined by \( \varepsilon_0 \varepsilon_r A/d \), where \( \varepsilon_0 \) is permittivity of free space \((8.85 \times 10^{-12})\), \( \varepsilon_r \) is relative permittivity \((3.9)\), \( A \) is unit area and \( d \) is the gate silicon oxide thickness \((3 \times 10^{-7})\). \( L \) and \( W \) represent the channel long \((50 \mu m)\) and wide \((25 \mu m)\), respectively. The effective hole mobility of the device in Figure 2a (main text) is calculated as follows:

\[ \mu = 10^4 \times \frac{I_d}{V_{gs}} \times \frac{L}{W} \times \frac{1}{C_{ox} V_{ds}} = 10^4 \times 1.22 \times 10^{-7} \times 2 \times \frac{1}{1.15 \times 10^{-4} \times 2} = 10.6 \text{ cm}^2/\text{Vs}. \]

**Measurements:** All electrical measurements were performed under ambient conditions using a Keithley semiconductor parameter analyzer, model 4200-SCS. XPS measurements were carried out by a Kratos AXIS-ultra spectrometer (UK) with the monochromatic Al K\( \alpha \) X-ray radiation \((1486.71 \text{ eV})\). The Raman spectra were performed in a WITec CRM200 confocal Raman microscopy system (laser wavelength 488nm and laser spot size is \(~0.5\mu m\)) and Si peak at 520 cm\(^{-1}\) was used as a reference for wavenumber calibration. Transmission electron microscope (TEM) was performed with a Hitachi JEOL JEM-2100F. Optical absorption measurements were performed in a Perkin Elmer Lambda 9 UV-Vis-NIR spectrometer.
**Figure S1.** Absorption spectra of the pristine and ACN-modified SWNTs deposited on Si substrates from the SWNTs after re-dispersing in a 2 wt % of co-surfactants (SDS: SC = 1:4 in weight). The initial weight ratio of SWNTs and ACN is 1:250.

![Absorption spectra](image)

**Figure S2.** (a) Output characteristics of the device as discussed in Figure 2a (main text). (b) The transfer curve of the same device, where the current is plotted in a linear scale.

![Output characteristics](image)
Figure S3. Relation between on-off ratio and hole mobility for the devices fabricated from ACN-functionalized SWNTs.
**Figure S4.** Transfer curves of a device prepared from ACN-modified tubes before and after vacuum annealing at 300°C for 2hr. It is noted that organic functional groups on SWNT surface can be removed by thermal annealing at above 300°C [So et al. *J. Am. Chem. Soc.* 2007, 129, 4866.]. If the ACN-modified metallic tubes are present in the conduction path, the metallic tubes shall be recovered and these devices are expected to become less gate-dependent after thermal annealing. The result here clearly demonstrates that the metallic tubes are still physically present in the networks.