# *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 $SiO_2$/Si substrate to form conducting channel ~50 μm long and ~25 μm wide. The gate dielectrics $SiO_2$

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 effecitve 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 ($8.85\times10^{-12}$), $\varepsilon_r$ is relative permittivity (3.9), A is unit area and d is the gate silicon oxide thickness ($3\times10^{-7}$ ). L and W represent the channel long (50 μm) and wide (25 μm), respectively. The effective hole mobility of the device in Figure 2a (main text) is cacluated as follows:

$$\mu = 10^4 \times \frac{I_d}{V_{gs}} \times \frac{L}{W} \times \frac{1}{C_{ox} V_{ds}} = 10^4 \times 1.22\times10^{-7} \times 2 \times \frac{1}{1.15\times10^{-4} \times 2} = 10.6 \text{ cm}^2/\text{Vs}.$$

**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 eV). The Raman spectra were performed in a WITec CRM200 confocal Raman microscopy system (laser wavelength 488nm and laser spot size is ~0.5μm) 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.

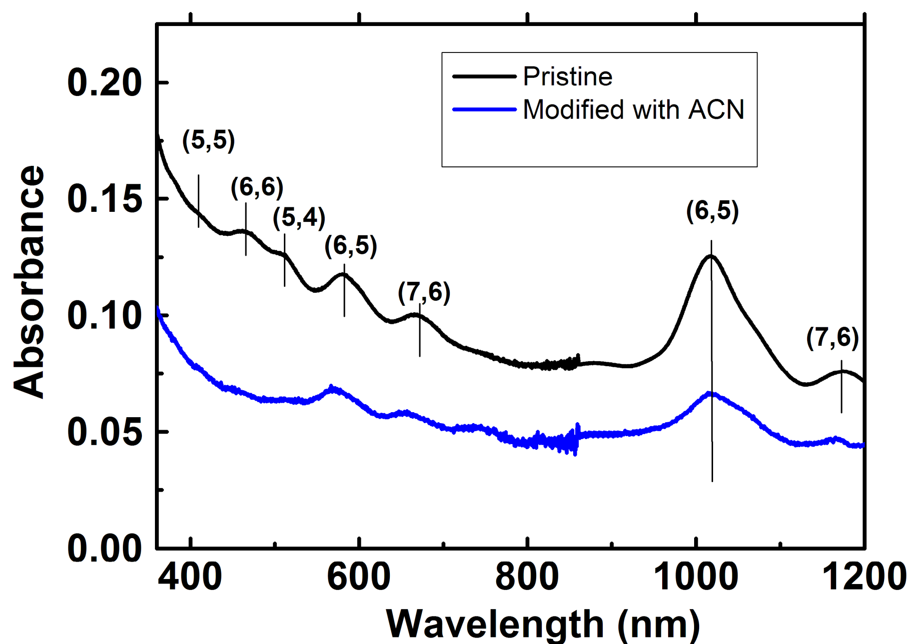
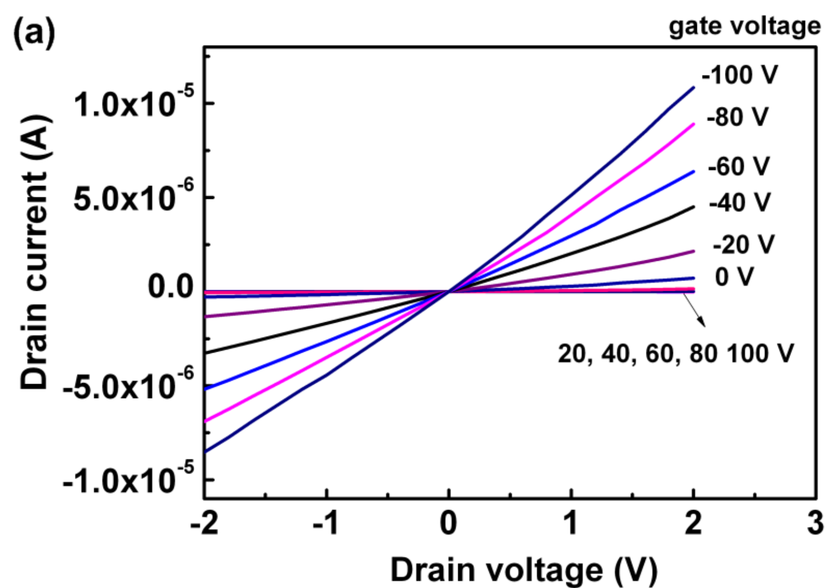



**Figure S2.** (a) Out put characteristics of the device as discussed in Figure 2a (main text). (b) The transfer curve of the same device, where the current is plotted in a linear scale.



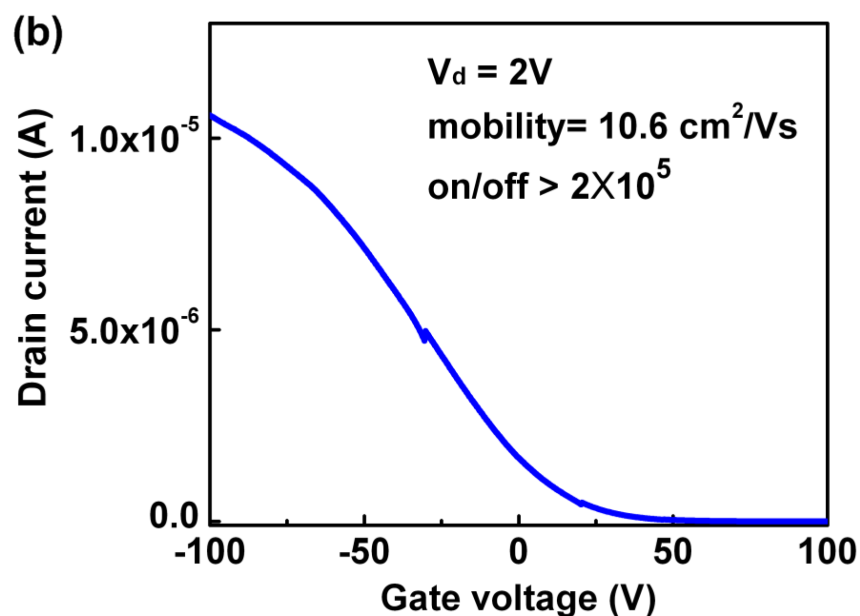



**Figure S3.** Relation between on-off ratio and hole mobility for the devices fabricated from ACN-functionalized SWNTs.

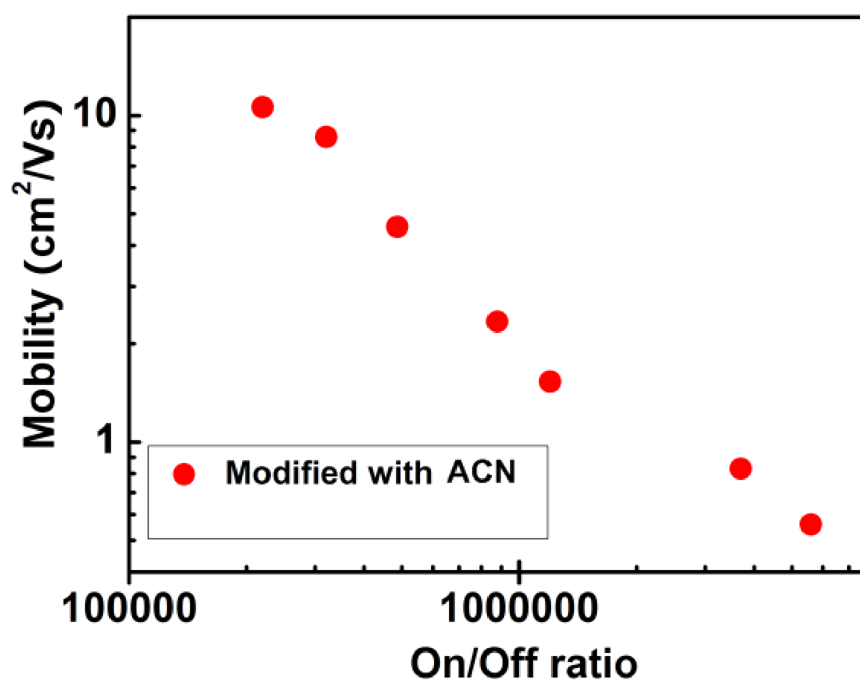


**Figure S4.** Transfer curves of a device prepared from ACN-modified tubes before and after vacuum annealing at 300°C for 2hr. It is noted that organic functional groups on SWNT surface can be removed by thermal annealing at above 300°C [So et al. *J. Am. Chem. Soc.* **2007**, 129, 4866.]. If the ACN-modified 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.