## Supporting Information

# Bulk Synthesis of Large Diameter Semiconducting Single-Walled Carbon Nanotubes by Oxygen-Assisted Floating Catalyst Chemical Vapor Deposition

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## 1. Experimental Details

### 1.1 Single-walled carbon nanotube growth

The single-walled carbon nanotubes (SWCNTs) were synthesized by oxygen-assisted floating catalyst chemical vapor deposition (FCCVD) at 1100 °C using $CH_4$ as carbon source. 500 sccm $H_2$ flowed through a quartz tube reactor with a diameter of 25 mm inserted into a horizontal tubular furnace. When the temperature of the reactor reached 1100 °C, a mixture of equal weights of ferrocene/sulfur powder

placed at the upstream of the reactor (where the temperature is 80°C) started to sublime and was transported into the reaction zone by the $H_2$ gas flow. Then, a flow of 3 sccm $CH_4$ and 0.2 sccm $O_2$ was introduced into the reactor. The synthesis process lasted for 60 min. Finally, the reactor was cooled to room temperature naturally.

### 1.2 Purification of the as-prepared SWCNTs

The as-prepared SWCNT thin films were heat treated at 200 °C in air for 2-5 h and then immersed in hydrochloric acid for 30 min. They were then thoroughly washed using deionized water and further annealed at 200 °C under an Ar flow for 30 min.

### 1.3 SWCNT thin film preparation

A filter was installed and connected to the tail gas pipe of the furnace used for SWCNT growth. Some of the SWCNTs synthesized by the FCCVD method were carried into the tail gas pipe by the $H_2$ carrier gas and were collected by the filter. By controlling the flow time, homogeneous SWCNT films with controllable thicknesses were obtained. We also collected SWCNT samples on a Cu grid for transmission electron microscopy observations using this method.

### 1.4 Absorption spectroscopy measurements

The as-synthesized SWCNT thin film was transferred from the filter onto the surface of a polished monocrystalline quartz film by contact and pressing. Then, the SWCNT film (together with the quartz substrate) was purified using the method described in part 1.2. The visible near-infrared (Vis/NIR) spectra (200–3000 nm) of the obtained SWCNT thin film was measured using a Varian Cary 5000 UV-vis-NIR spectrophotometer.

### 1.5 Fabrication and measurement of SWCNT film-based field effect transistors

Au electrodes with a channel distance of 2 □m were constructed on the surface of a 100-nm thick $SiO_2$/Si substrate. Then, the SWCNT thin film was transferred from the filter onto the surface of the substrate by contact and pressing. The purification of the SWCNT film was performed according to the procedure described in part 1.2. A gate voltage up to 5 V was applied to the underlying Si substrate, which served as the gate electrode to modulate the carrier concentration in the SWCNT network. A bias of 100 mV was applied between two neighboring electrodes that serve as the source and drain. The gate leakage current and the source-drain current were monitored and recorded. In all cases, the source-drain current significantly exceeded the gate leakage current. Sweeps of the gate bias were made from negative to positive bias. Hysteresis was observed depending on the sweep direction due to the presence of mobile charge, an effect routinely observed in SWCNT FET devices fabricated on 100 nm thick $SiO_2$ gate dielectrics.

## 2. Background subtraction of absorption spectra

The background absorption due to graphite, catalyst particle scattering and plasma absorption of carbonaceous material is assumed to be nonlinear which would better account for the physical features of the SWCNT absorption spectra such as peak overlap and transition broadening.[1] Basically, the background absorption is analyzed in two parts, which can be written as:[2]

$$A_{bg}(E) = kE^n + \Delta \tag{1}$$

where

$$\Delta = \frac{\omega_a (E_U - E\ ) + \omega_b (E_L - E\ )}{E_U - E_L} \quad . \qquad (2)$$

First the log-linear model is applied to simulate the nonlinear part $kE^n$ of the background absorption. Parameters $k$ and $n$ are empirical and determined at the region around 4 eV in the absorption spectra, where the π-plasmon absorption is expected to be dominant over the absorption due to electronic transitions from the SWCNTs. Then a small positive linear shift Δ of background and parameters in $A_{SWCNT}(E)$ are fitted. The remaining parameters $\omega = (\omega_a, \omega_b, \ldots, \omega_{n,m}, \ldots)$ are estimated based on absorption in the range from $E_L$=0.46 eV to $E_U$=2.50 eV. The SWCNT-related absorption spectrum is most identifiable in this range. Even if various compositions and properties of different bulk SWCNTs result in a deviation from the log-linear background model, we assume that the remaining background absorption is small and near-linear. The result of the above process is displayed in Figure 3b.

The weight ratios (r) of metallic tubes ($W_M$) and semiconducting tubes ($W_S$) are estimated from the corresponding absorption peak areas using the formula of: $r = W_M / (W_S + W_M)$. The weight ratio W is estimated from the absorption curves of metallic and semi-conducting tubes after the baseline subtraction. The r values of the 0-, 0.1-, 0.2-, and 0.3-SWCNTs are 0.30, 0.18, 0.12, and 0.17, respectively, indicating that effective enrichment of s-SWCNTs was achieved for the 0.2-SWCNT sample.

## References

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