

Electronic Supplementary Information

Preferential elimination of thin single-walled carbon nanotubes by iron etching

Fang Yu,^{ac} Haiqing Zhou,^{ab} Huaichao Yang,^{ab} Minjiang Chen,^{ab} Gang Wang^{ab} and
Lianfeng Sun^{*a}

^aNational Centre for Nanoscience and Technology, Beijing 100190, China.

^bGraduate School of Chinese Academy of Sciences, Beijing 100049, China

^cPeking University, Beijing, 100871, China

*Correspondence and requests for materials should be addressed to L. F. S.

(slf@nanoctr.cn)

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2. As-grown and thermally annealed Au-deposited SWNTs
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1. The synthesis, purification of the SWNTs film and Raman measurements

The SWNTs films used in our experiment were grown by catalytic floating chemical vapor deposition (CVD), with diameter distribution from 0.9 nm to 2.0 nm. To perform the iron cutting process, we firstly flattened the SWNTs on the surface of water and then transferred them onto the silicon substrate with a 300 nm SiO₂ layer, which was subsequently put on a hot plate and baked for about 1 h at 80 °C. 5 nm Fe film was thermally evaporated onto one end of the SWNTs film in a vacuum thermal evaporator at a deposition rate of 1.0 Å/s under a vacuum of $\sim 10^{-4}$ Pa. The catalytic cutting process was carried out at the center of a quartz tube in a tubular furnace. In order to activate the catalytic activity of Fe on SWNT films, the gas coflow of 400 sccm Ar and 70 sccm H₂ was applied, and the samples were thermally annealed at 850 °C for about 15 min to catalytic hydrogenation of carbon. After that, the samples were cooled down to room temperature under the protection of 100 sccm Ar. Scanning electron microscopy (SEM, Hitachi S4800), which can offer us a resolution of ~ 1 nm, was used to characterize the morphologies of Fe-decorated SWNTs at different regions.

The purification of iron-etched SWNTs was performed as follows: first of all, the iron-etched SWNTs were put into the tube furnace, and annealed at 250 °C for about 24 h under ambient conditions. Then the annealed samples were immersed into the solution of dilute hydrochloric acid for about 48h. Finally, the obtained samples were rinsed by deionized water repeatedly and then dried at 80 °C. After these processes, the samples were characterized by SEM and Raman spectroscopy.

In our experiments, the Raman measurements were carried out using a micro-Raman spectroscopy (Renishaw inVia Raman Spectroscope) under ambient conditions with three laser excitations, such as 514.5 nm (2.41 eV), 633 nm (1.96 eV) and 785 nm (1.58 eV). The laser power on the sample was set at around 1.0 mW to avoid laser induced heating. The application of a 100× objective lens with a numerical aperture of 0.90 can provide us a $\sim 1 \mu\text{m}$ laser spot size. Several spectra were collected to ensure the credibility and repeatability of the results.

2. As-grown and thermally annealed Au-deposited SWNTs

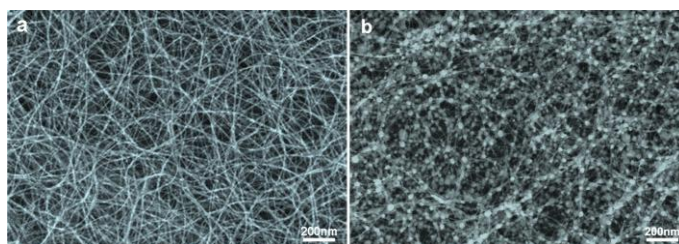


Fig. S1. As-grown (a) and Au (b) particles distributed on SWNTs after thermal annealing at the conditions same to that of iron etching process.

3. The regular alignment of suspended SWNTs after iron etching

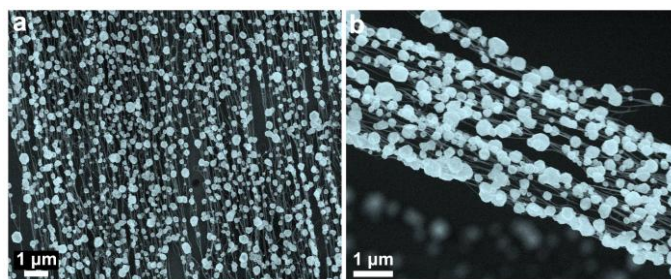


Fig. S2. The alignment of suspended SWNTs after iron etching, which may be related to the interaction between iron and SWNTs, and the movement of iron particles on the SWNTs' surface.

4. TEM analysis on the tube diameters of as-grown SWNTs

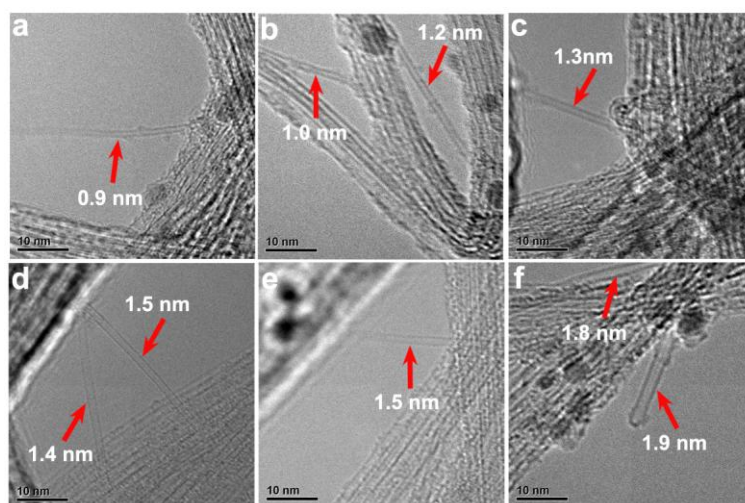


Fig. S3. TEM analysis of the tube diameters of as-grown SWNTs synthesized by catalytic floating chemical vapor deposition.

It is clearly found that the diameter of individual SWNT is in the range from 0.9 nm to 2.0 nm, which is calculated from Raman spectra of a film of SWNTs synthesized by catalytic floating chemical vapor deposition. This means that since there is a one-to-one correspondence between RBM peak frequency and tube diameter of individual SWNT: $\omega_{RBM} = 217.8 / d_t + 15.7$, Raman spectroscopy-based technique is an alternative method to that of TEM, which is also powerful and effective in determining the tube diameter of individual SWNT.

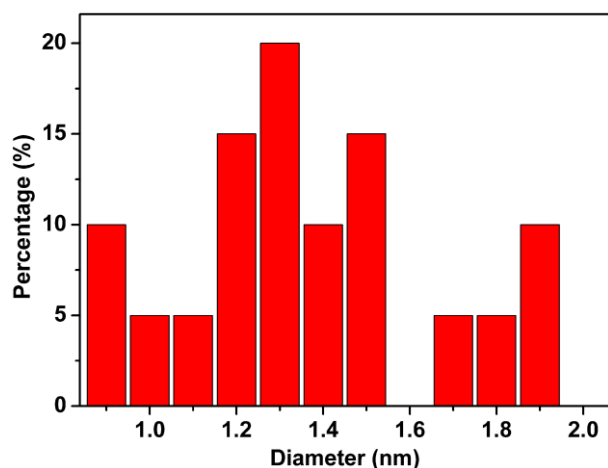


Fig. S4. Diameter distribution of as-grown SWNTs from totally 60 SWNTs characterized by TEM.

5. Raman spectra of iron-etched and 5 nm Fe-deposited SWNTs

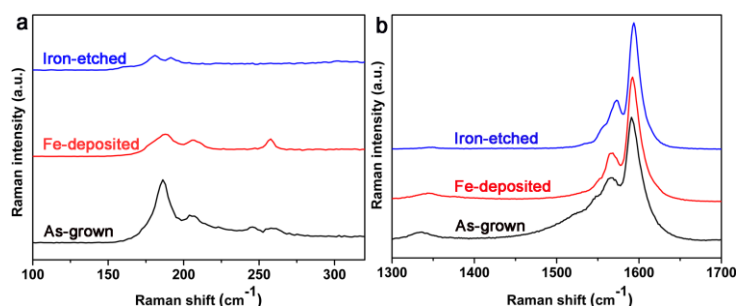


Fig. S5. The comparison among the Raman spectra of as-grown, 5 nm Fe-deposited and iron-etched SWNTs at 514 nm laser excitation.

For 5 nm Fe-deposited SWNTs, the RBMs and G mode in the Raman spectra are very similar to that of as-grown SWNTs. The frequencies of RBMs are still composed of lower and higher frequencies similar to those of pristine samples, which are in sharp contrast to our observed disappearance of RBM frequencies above 200 cm⁻¹ for iron-etched SWNTs. These observations also help us to conclude that iron etching can be applied to selectively remove thin SWNTs based on the catalytic hydrogenation of

carbon. At the same time, it is quite interesting to find that for the iron-etched SWNTs, the defect-related D mode is almost undetectable, meaning that external defects are not introduced by the technique based on iron etching.

6. Iron particles distributed on SWNTs film annealed without H₂ presence

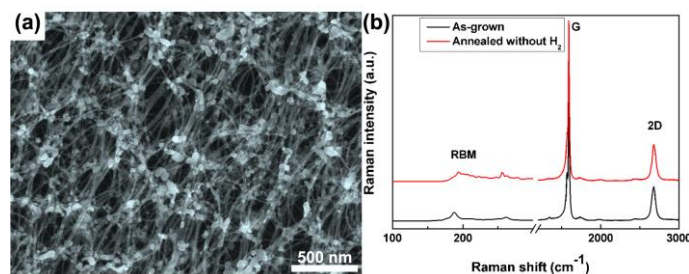


Fig. S6. (a) The typical SEM image of SWNTs film after similar iron etching without H₂ presence. (b) The corresponding typical Raman spectrum of iron particle-deposited SWNTs.

7. Typical SEM and Raman spectra of purified iron-etched SWNTs



Fig. S7. The typical SEM image of purified iron-etched SWNTs

According to the SEM image in Fig. S7, almost no iron nanoparticles can be observed. Also, some features have been found in Raman spectra of the samples after purification: firstly, the RBM peaks in the low frequency regions (100 cm⁻¹ – 300 cm⁻¹) of purified samples have few differences from that of iron-etched SWNTs (Fig.

3 and Fig. S8). Secondly, the defect-related D band (around 1350 cm^{-1}) is present with a slightly increased peak intensity, which may be induced during the purification.

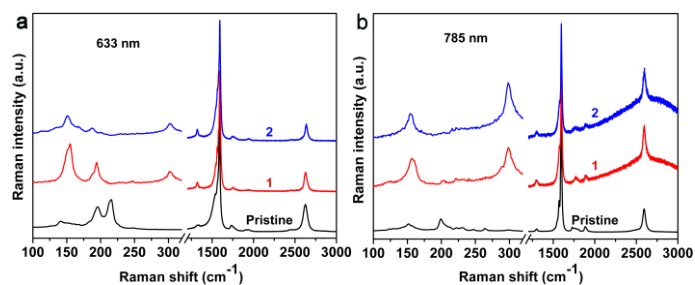


Fig. S8. The comparison of Raman spectra of purified (blue), iron-etched (red) and as-grown (black) SWNTs measured on region 1 and 2 at different laser excitations. (a) 633 nm and (b) 785 nm. The peak at around “ 300 cm^{-1} ” can be ascribed to the Raman signals of the silicon substrate which support the SWNTs.