Supporting Information for

Growth of Half-Meter Long Carbon Nanotubes Based on Schulz-Flory Distribution

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1. Supporting Figures

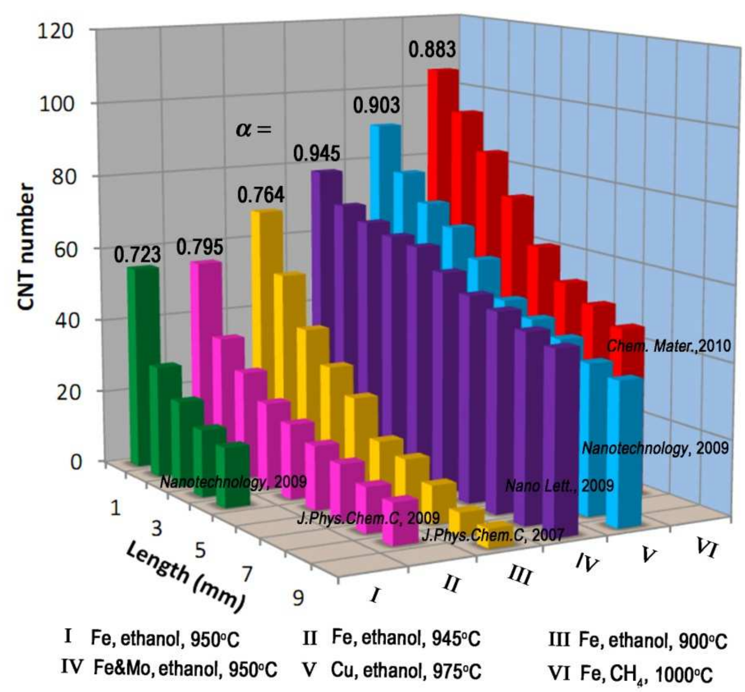

Figure S1. Number distribution of ultralong CNTs synthesized with different methods (I[1], II[2], III[3], IV[4],V[5], VI[6]). For instance, in method I, Fe nanoparticles works as the catalysts, ethanol is the carbon source, the growing temperature is 950 ^{o}C, and $\boldsymbol{\alpha}$ =0.723 refers to the corresponding catalyst activity probability.

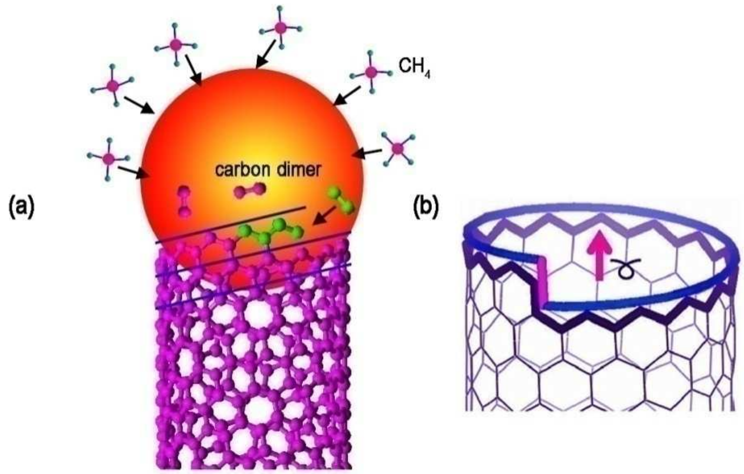

Figure S2. Illustration of screw-dislocation-like growth of CNTs. (a) Stimulation of growth of CNTs atom by atom. The growth of the CNT can be regarded as adding the carbon atoms dimer by dimer along the blue lines. (b) Illustration of an axial screw dislocation in a CNT (cited from referrence[7]).

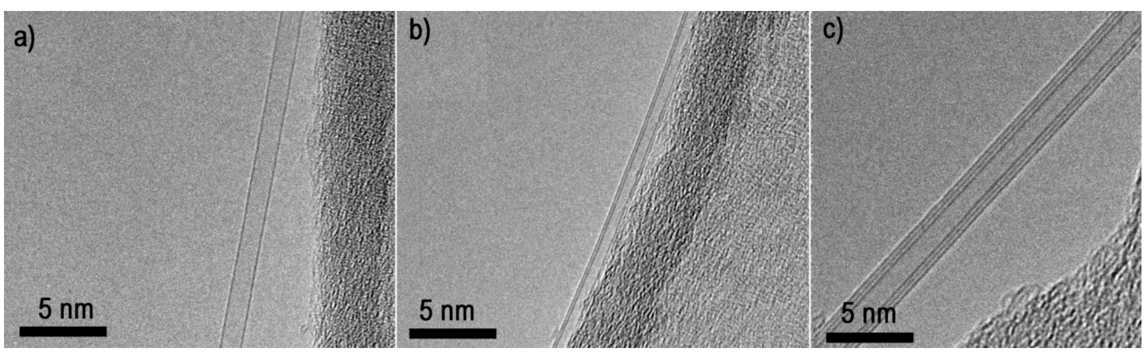

Figure S3. TEM images of as-grown ultralong CNTs. (a) Single-walled CNTs. (b) Double-walled CNTs. (c) Triple-walled CNTs.

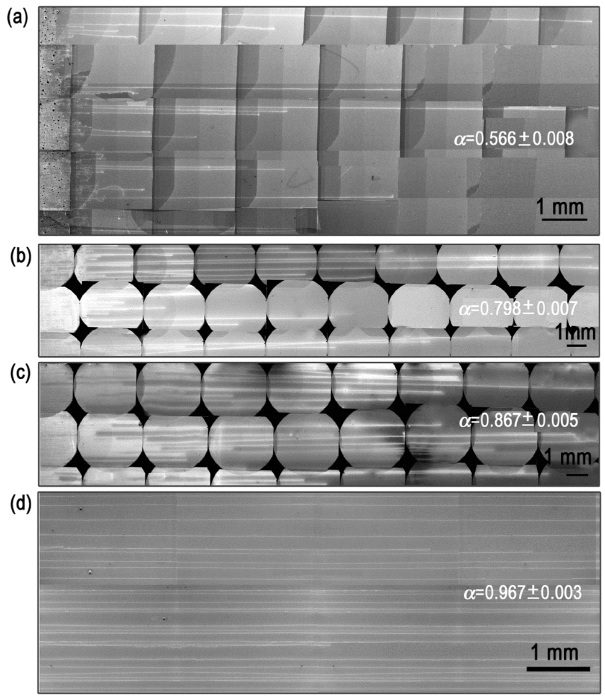

Figure S4. SEM images of ultralong CNTs with different number density distribution. (a) v=1.36 mm/s, T= 970^{o}C, c_{cat}=0.01 mol/L, W_{H2O}=0.3%, $R_{H2/CH4}$=1.8. (b) v=1.36mm/s, T=980^{o}C, c_{cat}=0.3 mol/L, W_{H2O}=0.3%, $R_{H2/CH4}$=2. (c) v=1.64mm/s, T=1000^{o}C, c_{cat}=0.5 mol/L, W_{H2O}=0.3%, $R_{H2/CH4}$=2.1. (d) v=1.58 mm/s, T=1010^{o}C, c_{cat}=0.5 mol/L, W_{H2O}=0.4%, $R_{H2/CH4}$=2.

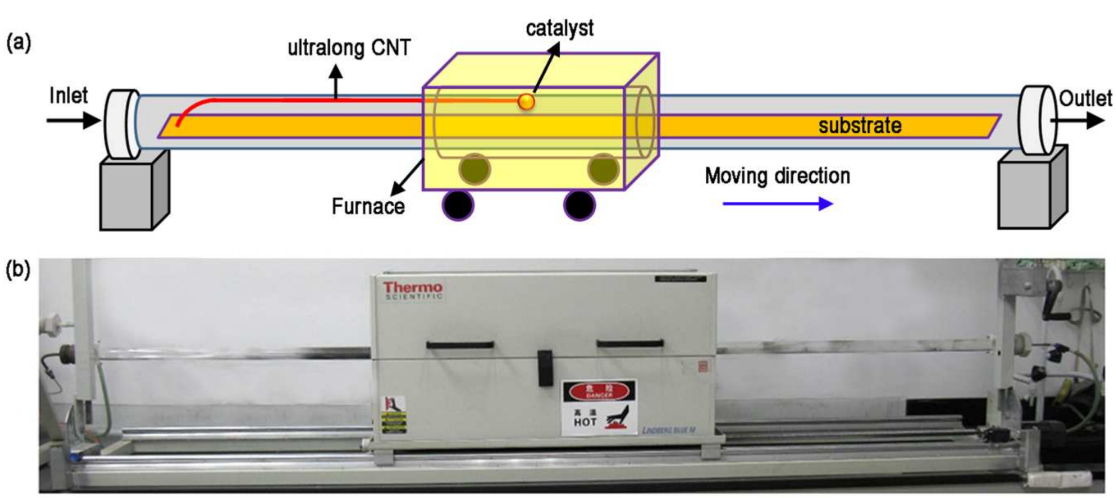

Figure S5. Furnace-moving method for synthesizing meter-long CNTs. (a) Schematic illustration of furnace-moving method. (b) Optical image of a moving furnace.

2. Supporting Texts

Supporting Text S1

Theoretical analysis of percentage and number density of ultralong CNTs

For a CNT with a length $\boldsymbol{L}$, the number of unit length can be regarded as $\boldsymbol{L}$. When the CNT grows to $\boldsymbol{(L-1)}$ unit lengths, the corresponding probability is $\boldsymbol{\alpha^{(L-1)}}$. After adding the $\boldsymbol{L^{th}}$ unit length, the CNT stops growing, which means that the catalyst is not active enough to maintain the CNT growth. The probability of catalyst losing its activity is $\boldsymbol{(1-a)}$. Thus the total probability for the CNT having a length $\boldsymbol{L}$ can be expressed as

$$P_L=\alpha^{(L-1)}(1-\alpha) \qquad (1)$$

The probability for the CNT having a length $\boldsymbol{L}$ is equal to the percentage of CNTs with length $\boldsymbol{L}$ in the total CNTs.

There is also a relationship between the number density of ultralong CNTs and catalyst activity probability. The number density can be defined as the total percentage of all the CNTs at a certain position, which is

$$d_L=P_L+P_{L+1}+P_{L+2}+\ldots\ldots+P_\infty \qquad (2)$$

$$=\sum_{L}^{\infty} P_L$$

$$=\sum_{1}^{\infty} P_L - \sum_{1}^{L-1} P_L$$

$$=\boldsymbol{S_{\infty}\text{-}S_{L-1}}$$

, where $\boldsymbol{P_L=\alpha^{(L-1)}(1-\alpha)}$ is a geometric progression and $\boldsymbol{S_L}$ is the sum of the geometric progression. According to the summation formula of geometric progression, we know that $\boldsymbol{S_{L-1}=(1-a)(1-a^{(L-1)})/(1-a)=1-a^{(L-1)}}$, while $\boldsymbol{S_{\infty}=(1-a)(1-a^{\infty})/(1-a)=1}$ (f*or* $0<\boldsymbol{a}<1$). Thus,

$$\boldsymbol{d_L=S_{\infty}\text{-}S_{L-1}=1\text{-}(1\text{-}a^{(L-1)})= a^{(L-1)}}$$

that is,

$$\boldsymbol{d_L= a^{(L-1)}} \qquad (3)$$

Supporting Text S2

Reason for the length distribution of vertically aligned CNT arrays cannot be expressed by SF distribution

For vertically aligned CNT arrays (VACNTAs), their growth speed is not constant. Generally, the VACNTAs follow base-growth mechanisms and their catalysts keep attached on the substrate. At the initial growing stage, the growth speed of VACNTAs is relatively high. However, the catalyst gradually deactivates due to the deposition of amorphous carbon on them and their growth speed slows down. This means that the growth is in an unstable state and the growth speed is not constant. The growth of

VACNTAs doesn't satisfy the prerequisites of SF distribution. Thus the length distribution cannot be interpreted by SF distribution.

Supporting Text S3

Calculation of catalyst deactivation probability for $\alpha_{mm} = 0.995$

The value of α is dependent on the selection of unit length, which can be 1 nm, 1 μm, 1mm, 1 cm and even 1 m. No matter for what unit length, the probability α based on one unit length can be definitely calculated from α based on another unit length, for instance, $a_{cm} = a_{mm}^{10}$. Because 1 mm= 1000000 nm, thus $\alpha_{mm} = \alpha_{nm}^{1000000}$. For a 1-nm-long single-walled CNT with diameter of 1 nm, there will be 60 carbon dimers in the CNT. Therefore, $\alpha_{nm} = \alpha_{\text{dimer}}^{60}$. Thus we obtain $\alpha_{mm} = \alpha_{\text{dimer}}^{60000000}$. When $\alpha_{mm} = 0.995$, we get the probability that the catalyst loses activity during the process that adding a carbon dimer to the CNT is $(1\text{-}\alpha_{\text{dimer}}) = 8.35 \times 10^{-11}$.

Referrences:

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