Acoustic-assisted assembly of an individual monochromatic ultralong carbon nanotube for high on-current transistors

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Great effort has been applied to scientific research on the controllable synthesis of carbon nanotubes (CNTs) with high semiconducting selectivity or high areal density toward the macroscale applications of high-performance carbon-based electronics. However, the key issue of compatibility between these two requirements for CNTs remains a challenge, blocking the expected performance boost of CNT devices. We report an in situ acoustic-assisted assembly of high-density monochromatic multivallent CNT tangles (m-CNT-Ts), consisting of one self-entangled CNT with a length of up to 100 mm and consistent chirality. On the basis of the minimum consumed energy model with a Strouhal number of approximately 0.3, the scale could be controlled within the range of $1 \times 10^4$ to $3 \times 10^6 \mu m^2$ or even a larger range. Transistors fabricated with one m-CNT-T showed an on/off ratio of $10^4$ to $10^6$ with 4 mA on-state current, which is also the highest on-state current recorded so far for single CNT–based transistors. This acoustic-assisted assembly of chiral-consistent m-CNT-Ts will provide new opportunities for the fabrication of high-performance electronics based on perfect CNTs with high purity and high density.

**INTRODUCTION**

Although carbon nanotubes (CNTs) are the most viable option as a substitute material for silicon for the next-generation transistors (1), the requirement of dense, aligned arrays of electronically pure CNTs is an enormous challenge for applications (2). Compared with post-treatment synthesis (3, 4), the in situ synthesis of CNTs with specific structures is a direct and nondestructive method, in which the composition and dispersion of catalysts have been demonstrated to be of central importance in controlling the density (5), semiconducting selectivity (6), and chirality (7). However, it is scarcely possible to directly synthesize pure semiconducting CNTs by an in situ catalytic reaction only, without any post-separation, and small amounts of metallic CNTs may cause catastrophic device shorting failures. Thus, the synthesis of CNTs with consistent chirality and high density is facing a bottleneck, and novel technical routes should be explored to further develop the field of carbon-based electronics.

Most of us have been incautiously trapped by the technical standards proposed by IBM in 2013 that have singled out the necessity of a high density of up to $125 \text{nm}^{-1}$ in tandem with a high semiconducting selectivity of up to 99.9999% of CNT arrays for a larger output current and a lower energy consumption in electronics (2). However, an important prerequisite for single-walled CNTs (SWNTs) with an average diameter of 1 nm was ignored, and few-walled CNTs, especially double-walled CNTs (DWNTs) and triple-walled CNTs (TWNTs), were not well received because of the challenge of the reproducible synthesis of all-semiconducting few-walled CNTs. If any inner wall of an individual few-walled CNT is metallic, it will lead to leakage currents and a decreased on/off ratio in CNT field effect transistors (CNTFETs). Recent experimental reports have demonstrated that ultralong CNTs synthesized with the assistance of water were mostly all-semiconducting DWNTs or TWNTs and that a representative TWNT with a perfect structure could withstand the maximum current of 17 mA, which is six times higher than that of SWNTs (8), and exhibited sensitive light responses (9). Considering the standard proposed by IBM based on the SWNT arrays with an average diameter of 1 nm, a density of up to 125 SWNTs $\mu m^{-1}$, and an output current of up to 3 $\mu A$ per CNT, the total current drivability within the 1-$\mu m$ range reaches 0.375 mA. In contrast, for TWNTs with an average diameter of 3 nm and an output current of up to 17 $\mu A$ per CNT, the density will decrease astonishingly to 22 TWNTs $\mu m^{-1}$ for the same current drivability, markedly shortening the path toward the ultimate goal. On the other hand, half-meter-long CNTs with an atomically consistent perfect structure and superior properties could be synthesized using the furnace-moving method (10); however, the as-synthesized horizontally aligned ultralong CNTs usually have different chiralities and a tip-growth mode determined low density, resulting in difficulties in effective separation. Thus, it will be easier to achieve the most difficult step of short CNT separation and manipulation for further applications if we assemble ultralong one-dimensional (1D) chiral-consistent CNTs into macroscopic quasi-planar structures with a considerable density.

In actuality, tangling is a common phenomenon according to some related research on elastic rods immersed in glycerin (11), silk filaments in a flowing soap film (12), and flagella of swimming bacteria (13) derived from fluid-body interactions. Furthermore, our previous studies showed that a coupling of the flow and a sound field could break the equilibrium state between the low-velocity gas flow and the ultralong CNT (14). These results suggest that the in situ assembly from an entangled ultralong CNT to a large-area planar structure through the means of a sound field is feasible. We believe that this assembly of 1D electronically pure nanotubes will provide new approaches and routes for fabricating high-performance carbon-based electronics.

**RESULTS**

In situ assembly of monochromatic CNT tangles

Figure 1A shows a schematic illustration of the synthesis of monochromatic CNT tangles (m-CNT-Ts) by introducing acoustic waves...
in situ during the normal growth process of ultralong CNTs (see Materials and Methods for details). The synthesis of ultralong CNTs via the chemical vapor deposition method relies on many factors, such as temperature, gas velocity, composition of reactants, and gas flow (10), of which gas flow stability is important for the control of the horizontally aligned morphology. Therefore, we specifically designed a layered rectangular reactor (LRR) with a width-to-height aspect ratio of 120:12 (fig. S1), the structure of which led to a reduction of velocity fluctuation along the height direction and a significantly stabilized gas flow during the reaction. The gas flow will therefore be very sensitive to external interference, leading to easy entanglement of some floating ultralong CNTs under the effect of acoustic-assisted gas flow before the ultimate formation of the whole m-CNT-T shown in (B). The loop diameter distribution was counted as shown in (D). Scale bars, 20 μm for (B) and 1 nm for (C).

**AIV mechanism for the formation of m-CNT-Ts**

The growth flow for the formation of m-CNT-Ts in the LRR is a plane Poiseuille flow (17), in which the initial parabolic pattern (fig. S3) was abruptly disturbed with a periodic jet driven by a loudspeaker (fig. S4) from a wall orifice, and thus, velocity fluctuations in tandem with vortices were formed. Then, the vortices would develop under the effect of viscous diffusion and pressure distribution (18). Because of the friction against the wall, the pressure gradient is smaller at the inlet and gradually increases toward the outlet (19). Thus, the forward vorticity is lower than the backward, which denotes the morphology of m-CNT-Ts with a straight segment in the upstream. As the vortices propagated downstream, they combined into larger vortices and interacted with each other, stretching and rotating and eventually consuming energy to form smaller vortices. Because the interaction was spatially symmetric, these small vortices became isotropic, with a round shape as the secondary structure. Thus, the size of the secondary formed nanotube loops may be associated with the minimum size of the vortex (see Supplementary Text for a detailed analysis).

To explore the principle of the formation of m-CNT-Ts and to test the working mechanism described above, we synthesized m-CNT-Ts at two different optimized gas velocities, determined by our previous work (10), for various acoustic wave frequencies. Depending on the growth conditions, the resultant m-CNT-Ts could be relatively large and dense or relatively small (area = 1 × 10^4 to 3 × 10^4 μm^2; fig. S2). Furthermore, we performed a demonstration experiment with a hot wire to visualize the phenomenon of AIVs (movie S1) and established a mathematical model for a floating CNT under an acoustic wave to provide a method for further calculations (see Materials and Methods for details). Moreover, the relationships between the acoustic wave frequency (f), gas flow velocity (u), and average diameter of the secondary nanotube loops (D) were established using a Strouhal number (St; a dimensionless parameter) model, St = fD/μ, which was used to describe the tail or wing kinematics of swimming and flying animals (12, 20–22). Natural selection is expected to favor high propulsive efficiency with minimally consumed energy over a narrow range of St, which usually peaks within the interval 0.2 < St < 0.4 (21). As shown in Fig. 2B and table S1, the m-CNT-Ts follow a similar natural selection, as shown by the as-defined St, which also exhibits distributions within the same range. Thus, the minimum dissipative energy principle in nature seems to work even at the nanoscale. Therefore, the average scale of an m-CNT-T can be optimized by adjusting the frequency of the acoustic wave under given growth conditions, as indicated in Fig. 2C. It is important to note that the frequency must be confined to a narrow range between 10 and 40 Hz because a higher frequency may cause CNT fracture, whereas a CNT may still be straight at a lower frequency (fig. S5). Atomic force microscopy characterization also demonstrated the CNT fracture because there was no catalyst particle at the end of the CNT (fig. S6). Additionally, according to the Schuld-Flory distribution, for the growth of ultralong CNTs with the catalyst activity probability above 92%, CNTs longer than 15 cm will be reduced by 66.7% compared to the number of those grown in the catalyst region (10). The sparse arrays of long nanotubes with their pitch widely distributed in the 2- to 6-mm range can ensure that each as-grown

**Fig. 1. Synthesis of m-CNT-Ts.** (A) Schematic illustration of the synthesis of m-CNT-Ts assisted with acoustic waves. For chiral characterization, under the illumination of a supercontinuum laser, m-CNT-Ts of different structures show different colors. Unchanged color for each ultralong CNT indicates that the chirality is consistent. (B) Scanning electron microscopy (SEM) characterization of a representative as-grown m-CNT-T (growth condition: f = 30 Hz, u = 1.7 mm s\(^{-1}\)). (C) Cs-corrected TEM image of a triple-walled ultralong CNT. An individual ultralong CNT will curl to form many small loops under the effect of acoustic-assisted gas flow before the ultimate formation of the whole m-CNT-T shown in (B). The loop diameter distribution was counted as shown in (D). Scale bars, 20 μm for (B) and 1 nm for (C).
m-CNT-T with its width of up to hundreds of micrometers is made of only one ultralong CNT and will not entangle with other CNTs nearby, which is a prerequisite to producing single-chirality m-CNT-T. The pitch distances increase with the increasing length of the ultralong CNTs, providing a greater likelihood of producing larger m-CNT-Ts.

Chiral consistency verification of ultralong CNTs and m-CNT-Ts

In addition to its effect of stabilizing the gas flow, LRR provides a facile route for the large-scale synthesis of ultralong CNTs. By controlling the gas velocity distribution carefully and enlarging the invariable temperature area (fig. S1), five 100-mm wafers of ultralong CNTs were synthesized simultaneously in an LRR heated in an enclosed muffle furnace with hearth dimensions of 1000 × 300 × 300 mm³. These wafer-scale ultralong CNTs could be visible even by the naked eye under ambient conditions assisted with condensed vapor (23) (Fig. 3B and fig. S7), avoiding the size constraints for the samples under a standard electron microscope.

The wafer-scale synthesis of ultralong CNTs will make it easier to directly and widely identify the chiral structures with optical characterization methods. For chiral identification of CNTs, resonance Rayleigh scattering (RRS) is significantly more accurate and effective compared to fluorescence excitation spectroscopy and Raman spectroscopy (24). A structure-property “atlas” for SWNT optical transitions (24) has been established, and real-time true-color imaging of SWNTs (25) was also achieved, whereas RRS of few-walled CNTs can be more complicated because of the interaction between the walls (26). A 100-mm-long mono-chromatic ultralong CNT was observed under an optical microscope by wide supercontinuum laser illumination (Fig. 3, A and B), and mono-chromatic serpentine CNTs with 11 segments, formed by one 400-μm-long ultralong CNT, show no color change or resonance peak shift, signifying the consistent chirality (fig. S8, C and D) (25). This is the first time that monochromatic CNTs with decimeter lengths have been observed under a microscope. CNTs with different colors and chiral structures are shown in Fig. 3 (C and D) and fig. S8 (A and B). Raman spectra

Fig. 2. AIV growth mechanism of m-CNT-Ts. (A) Representative statistical data for the diameter distribution of small loops composing each m-CNT-T. The left panel shows diameter distributions for corresponding labeled m-CNT-Ts on the right panel. These m-CNT-Ts were synthesized under different conditions. Top to bottom: f = 35 Hz, u = 1.2 mm s⁻¹; f = 25 Hz, u = 1.7 mm s⁻¹; f = 35 Hz, u = 1.7 mm s⁻¹. (B) Statistics about Strouhal number for m-CNT-Ts synthesized under different conditions and other natural things. f1 to f5: from 15 to 35 Hz in the 5-Hz step, u1 = 1.2 mm s⁻¹, u2 = 1.7 mm s⁻¹. For example, f1 u1 means that the experimental condition is f = 15 Hz, u = 1.2 mm s⁻¹. Two dot-dashed lines indicate that St values for every m-CNT-T lie in the same interval as the reported natural things, and the dashed line indicates the reported optimal St. (C) Control law between the frequency of acoustic wave and the diameter of secondary nanotube loops. Three blue lines represent operation at u = 1.2 mm s⁻¹, whereas red lines represent operation at u = 1.7 mm s⁻¹. Scale bars from top to bottom in (A), 50, 50, 100, and 30 μm.
further demonstrated that each monochromatic ultralong CNT exhibited the characteristics of perfect structure and consistent chirality. Double radial breathing mode (RBM) peaks shown in Fig. 3I indicated that this may be a DWNT, in agreement with the previously reported results \( (27) \). Moreover, if we consider the proposed standard \( (2) \) on a wafer scale, SWNT arrays with a density of up to 125 SWNTs \( \mu m^{-1} \) and an average length of 7 nm weigh 2.35 \( mg \) when closely arranged on a 100-mm silicon wafer. For TWNTs with an average length of 10 cm and a diameter of 3 nm, the calculated density will decrease to 21 TWNTs \( \mu m^{-1} \) for the equivalent mass. A recent breakthrough has demonstrated that we are closer to this target through the “in situ catalyst loading” approach \( (28) \).

Additionally, because of the polarization of CNTs, a complete picture will not be observed if the CNT is curved, as shown in the bottom panel of Fig. 3A. Nevertheless, we could obtain a nearly complete image for an m-CNT-T (Fig. 4B) by rotating the objective table (Fig. 4A) and combining snapshots taken from different angles. A red m-CNT-T was thus observed completely (Fig. 4C; see original pictures in fig. S9), although some invisible CNT segments were evident through the comparison with the corresponding SEM image (Fig. 4D). Raman spectra recorded from four different positions of the m-CNT-T with 633-nm excitation (Fig. 4E) show a hardly noticeable D band; however, the same RBM bands shift, indicating that the m-CNT-T is of high quality with consistent chirality. Furthermore, RRS spectra with the same resonance peak shifts (Fig. 4F) are clear evidence of the consistent chirality \( (25) \).

**Photoelectronic properties of m-CNT-Ts**

To evaluate the performance of m-CNT-Ts, we fabricated back-gated field effect transistors with a representative schematic structure, with the SEM images shown in Fig. 5 (A and B). The transfer characteristics \( (I_D - V_G) \) measured at various \( V_D \) biases are shown in Fig. 5C, indicating that the device consists of an all-semiconducting m-CNT-T. Moreover, these transistors exhibit an on/off ratio above 1000 and deliver a respectable on-state current of 4.4 mA at \( V_D = 2 \) V and \( V_G = -20 \) V, showing promising compatibility between the high on/off ratio and high output current in CNTFETs. Meanwhile, this is also the highest output current recorded to date for single CNT–based transistors. The output characteristics \( (I_D - V_D) \) of the same device shown in Fig. 5D also reveal the
formation of ohmic contacts between the m-CNT-T and the metal contacts. Furthermore, a sensitive photo response of the m-CNT-T to short-wave infrared is also demonstrated by the photocconductance spectra shown in Fig. 5E, implying that the chirality is consistent and that the m-CNT-Ts show great potential for use in optoelectronic devices. Furthermore, because of the special morphology of m-CNT-Ts with a straight segment next to the tangle, the areal density of m-CNT-Ts can be accurately and efficiently evaluated on the basis of the ratio of current intensity between the tangle and the straight segment only if the CNTs’ wall number has been confirmed.

Contrary to the previous prejudice against the use of few-walled CNTs in electronics, transistors based on few-walled ultralong CNTs exhibit superior performance with an on/off ratio up to 10^7 and a higher output current than that in the usual SWNT-based devices (Fig. 5, F and G) (29). Additional representative measurement results indicate that the on/off ratios range from 10^3 to 10^7, whereas the output current can easily exceed 15 μA (fig. S10), which corresponds to a high semiconducting ratio of ultralong CNTs of up to 92.6% (fig. S11). The devices shown in Fig. 5B based on m-CNT-Ts demonstrate a lower on/off ratio that is believed to be due to the screening effect between the CNTs or the dirty surface of substrates. More CNTFETs fabricated with m-CNT-Ts shown in fig. S12 demonstrate a higher on/off ratio of up to 10^5 to 10^6 at lower V_D biases. Thus, devices based on these m-CNT-Ts have achieved the compatibility of the considerable current density and on/off ratio that is more competitive than those of most existing CNT products (3, 5, 16, 29, 30) and have exhibited a promising potential for applications in logic circuits and photodetector or radio frequency circuits (Fig. 5H). As the next step, we will synthesize more and larger m-CNT-Ts directly on silicon wafers; this will not only avoid the dirty matter originating from the cutting of the wafers but will also contribute to the separation and applications of m-CNT-Ts at a larger scale. Furthermore, the controlled synthesis of longer CNTs with reduced diameters is another key factor because the longer length probably ensures the all-semiconducting behavior through an individual few-walled CNT; additionally, a reduced diameter may have a positive effect on the turn-off performance of the transistors fabricated using a thin gate dielectric. We believe that further steps must be taken to explore and improve the device performances based on m-CNT-Ts.

**DISCUSSION**

Extremely high requirements have been proposed for carbon-based electronics. The areal density of aligned CNTs can reach 130 CNTs μm⁻² through a catalytic reaction (5) or 500 CNTs μm⁻² by post-assembly (3). Thus, the density standard has not been the key problem, and it can even be lowered by six times to 22 CNTs μm⁻² for SWNTs. However, there exists an unavoidable obstacle that if the sample contains more than 0.0001% metallic CNTs, the as-fabricated devices will face shorting failure. Thus, it is necessary to explore new technical routes. The initial aim of assembling every decimeter- to meter-long CNT into single-chirality macroscopic tangles is aimed at making it easier to separate and manipulate CNTs on the basis of RRS. However, this is only the first step of our road map for wafer-scale applications of m-CNT-Ts. We can synthesize more and larger m-CNT-Ts directly on wafers so that the separation can be performed on a larger scale because of the advantage of wafer-scale synthesis capability within the LRR. Then, m-CNT-Ts with the same color can be enriched on the designated substrate, even covering the whole substrate for ideal film formation. Because this film theoretically shows single chirality and superlong length, transistors or other devices fabricated on it are most likely to exhibit high performance. Furthermore, if we succeed in aligning these enriched m-CNT-Ts on wafers, the CNTFET performances will achieve another leap. Therefore, whereas the long-pursued CNTFET application is demanding and ultimately many technological barriers will have to be overcome, the proposed method offers another approach circumventing the purity and density problems, although the placement and uniformity problems need to be further addressed in future studies.

In summary, we proposed an in situ assembly approach for the synthesis of large-area (1 × 10⁴ to 3 × 10⁴ μm²) quasi-planar m-CNT-Ts from decimeter-long CNTs with consistent chirality. This large-scale
m-CNT-T was assembled by introducing acoustic waves into the growth system of ultralong CNTs, and an innovative AIV mechanism was proposed for the formation of m-CNT-Ts. The scale of m-CNT-Ts could be controlled on the basis of a natural selective model of minimum consumed energy with a Strouhal number of approximately 0.3. Furthermore, five 100-mm wafers of ultralong CNTs were first synthesized simultaneously in our homemade LRR and were visualized directly by the naked eye under ambient conditions assisted with condensed vapor. Additionally, chiral-consistent decimeter-long monochromatic CNTs and entangled CNTs were observed under a standard microscope based on RRS, with the Raman spectra further demonstrating chiral consistency and structural perfection. CNTFETs fabricated with the large-area m-CNT-Ts and entangled CNTs exhibited unique photoelectronic properties, showing a feasible method for compatibility between high semiconducting purity and high areal density for CNTs. In addition to their direct use in the fabrication of chips or devices, these CNTs can also be stretched into a dense CNT array using the standard spinning treatment for millimeter-long fibers (15), offering a new route for high-density flexible electronic materials. Finally, we believe that the formation of a specific architecture assisted by AIVs is also feasible for other flexible materials synthesized in a gaseous environment. Additional techniques will be explored to increase the density and area of these m-CNT-Ts, ultimately achieving effective separation based on RRS and large-scale applications in electronics, transparent display, sensors, superstrong fibers, aeronautics and astronautics, and even space elevators.

MATERIALS AND METHODS
Synthesis of m-CNT-Ts and serpentine CNTs
m-CNT-Ts were synthesized in our designed LRR heated in customized muffle furnace (made by CNT Furnace Co. Ltd.). FeCl₃ ethanol solution (0.03 M) was deposited as a catalyst precursor onto silicon substrates with an 800-nm-thick SiO₂ coating layer. The temperature was increased at a rate of 19.5°C min⁻¹ at an atmosphere of H₂/Ar and kept at 1005°C for 15 min. Then, CH₄ and H₂ (V₁ = 20 V, V₂ = -20 V, V₃ = 0 V, V₄ = 0 V, V₅ = 40 V) were inlet into the reactor continuously for 30 min (10). When t = 20 min during reaction, an acoustic wave (f = 25 Hz) generated by a signal generator (RIGOL DG1022U, 1 μHz RF, radio frequency).
to 25 MHz for sine wave, <40 W) was introduced into the reactor for the last 10 min.

The procedure for synthesizing serpentine CNTs was similar to that described above except without acoustic wave. \( T = 1005^\circ\text{C}, \mu = 1.7 \text{ mm s}^{-1}, C_{\text{cat}} = 0.03 \text{ M}, w_{120} = 0.46\%, \) and \( R_{12/14} = 2.06. \) Another difference was that the substrates were quartz having been annealed in air for 8 hours.

### A mathematical model about floating CNT under acoustic wave

According to the propagation feature of longitudinal wave, when acting as a simple harmonic force at one position \( x = 0 \) on the CNT, the vibration would transmit along the CNT. Suppose the deformation at \( x \) position and \( t \) moment was \( \delta(t, x) \), and at \( x + dx \) position (\( dx \) denotes a minimum vibration) was \( \delta(t, x + dx) \); thus, the total deformation was

\[
\delta(t, x + dx) - \delta(t, x) = \frac{\partial \delta(t, x)}{\partial x} dx
\]

Suppose the deformation was within elastic range, and according to the law of the elastic

\[
\frac{F_x}{S} = -E \frac{\partial \delta x}{\partial S} = -E \frac{\partial (\delta x + dx)}{\partial x}
\]

Drag force acting on the CNT \((31)\) was

\[
F_D = \frac{1}{2} \sqrt{2\pi m_k T}N \sqrt{\frac{4S}{\pi}} t^{1.1} V dx
\]

\[
= P \sqrt{\frac{4S}{\pi}} V dx \left( P = \frac{1}{2} \sqrt{2\pi m_k T} \Omega_{(1,1)} \right)
\]

Thus, the resultant force acting on the CNT was

\[
dF_x = F_x - F_{x+dx} - F_D = -\frac{\partial F_x}{\partial x} dx - F_D = SE \frac{\partial^2 \delta}{\partial x^2} dx - P \sqrt{\frac{4S}{\pi}} V dx
\]

According to Newton’s second law

\[
SE \frac{\partial^2 \delta}{\partial x^2} dx - P \sqrt{\frac{4S}{\pi}} V dx = \rho S dx \frac{\partial^2 \delta}{\partial t^2}
\]

Boundary condition was \((L \) denoted length of CNT)

\[
\left( \frac{\partial \delta}{\partial x} \right)_{x=0} = -\frac{F_x}{ES} \sin \omega t
\]

\[
\delta_{x=L} = 0
\]

### Characterization of m-CNT-Ts and serpentine CNTs

The as-grown samples were inspected with SEM (JSM 7401F, 1.0 kV), a Raman spectrometer (Horiba HR 800, 532/633 nm), and TEM (JEM 2010, 120.0 kV) to characterize the morphology and structure. An optical microscope (long working distance metallography microscope, FS 70Z) and a supercontinuum laser were used for RRS.

**Visualization of wafer-scale ultralong CNTs**

A purchased humidifier was reformed with a metal joint and a hosepipe to directly produce vapor. In a dark room with a lamp lighting nearby, the vapor was blown to the surface of a silicon substrate, and CNTs were visible with the naked eye. A bottle of liquid nitrogen was put under the substrates to maintain the image of ultralong CNTs to be shot with a micro lens. This method was also a further development derived from the reported vapor condensation-assisted optical microscopy.

**Fabrication and measurements of electronic devices**

Palladium (Pd) electrodes (70 nm thick) were patterned on Si/SiO2 substrates to maintain the image of ultralong CNTs to be shot with a microscope (long working distance metallography microscope, optical microscope (532/633 nm), and TEM (JEM 2010, 1200 kV). The procedure for synthesizing serpentine CNTs was similar to that described above except without acoustic wave. \( T = 1005^\circ\text{C}, \mu = 1.7 \text{ mm s}^{-1}, C_{\text{cat}} = 0.03 \text{ M}, w_{120} = 0.46\%, \) and \( R_{12/14} = 2.06. \) Another difference was that the substrates were quartz having been annealed in air for 8 hours.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/2/11/e1601572/DC1

### REFERENCES AND NOTES


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