Direct Growth of Single-Walled Carbon Nanotubes without Metallic Residues by Using Lead as a Catalyst

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The IVA group metal Pb was found to be an efficient catalyst for the chemical vapor deposition (CVD) growth of single-walled carbon nanotubes (SWCNTs) under suitable conditions. Especially the volatility of Pb made it feasible to realize the direct growth of SWCNTs with no metallic catalyst residues. Three methods including a low gas flow process, a fast-heating process, and a polymer-assisted process were used to control the volatility of lead at a suitable level. Both random SWCNT networks and horizontally aligned SWCNT arrays were efficiently grown on silicon wafers. The density of the SWCNT arrays can be altered by the CVD conditions. The electrical transport measurements for single tubes show that the produced SWCNTs are of high quality. These directly prepared SWCNTs with no metallic impurities will greatly benefit the study of the intrinsic properties of SWCNTs and are very important for making use of SWCNTs in nanodevices and in biological systems.

1. Introduction

The catalyst plays an important role in the chemical vapor deposition (CVD) process of single-walled carbon nanotubes (SWCNTs).[1] It is generally accepted that the nanoscaled catalyst particles act as the initiating centers and the SWCNTs grow out from them via a vapor−liquid−solid (VLS) mechanism.[2] Fe, Co and Ni are commonly used catalysts. Experimental[3,4] and theoretical[5−7] studies all show their high catalytic activities. However, there is still the motivation to explore new catalysts for the growth of SWCNTs in order to achieve the controllable synthesis of SWCNTs and/or to obtain SWCNTs of high purity for studying their intrinsic properties.

It was found that the types of catalysts could affect the diameter distribution of SWCNTs.[8] Therefore, the exploration of new catalysts for CVD preparation of SWCNTs will be helpful for both controllable growth of SWCNTs with specific diameters or chirality and in-depth studies of growth mechanism of SWCNTs. Recently, Cu was proved to be an efficient catalyst in our laboratory, which showed very high catalytic activity for the growth of both random SWCNT networks and horizontally aligned SWCNT arrays.[9] Noble metals such as Au, Ag, Pt, and Pd were also found to be able to catalyze the growth of SWCNTs.[10,11] These results inspire further exploration on new catalysts for SWCNTs' growth.

SWCNTs obtained using common transition metal catalysts such as Fe, Co, and Ni often have residual ferromagnetic metallic impurities. This is a big problem for the study of the intrinsic properties of SWCNTs and the application of SWCNTs in some areas. For example, these metallic impurities may suppress the SWCNTs' field electron emission,[12] obscure or alter SWCNTs' magnetism,[13] and affect electrical transport properties[14] and electronic behavior of SWCNTs.[15] The purification procedures to eliminate the residual catalysts and other impurities can introduce defects into the SWCNTs. Therefore, to directly prepare SWCNTs containing no residual metal catalyst is a big challenge.

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In this work, we report that the IVA group metal Pb can be an effective catalyst for CVD growth of both random SWCNT networks and horizontally aligned SWCNT arrays on silicon wafers. As the melting point of Pb is quite low (327 °C), the Pb catalyst volatilizes continually in the growth process and eventually disappears at the end of the growth. This offers us a way to directly prepare SWCNTs with no metallic impurities. These SWCNTs containing no metal catalyst can be used to study the intrinsic properties of SWCNTs. They can also be applied in nanodevices[15] and in biological systems,[16] where metallic impurities are also harmful.

Pb was chosen as a potential catalyst for SWCNTs growth because the binary phase diagram of C−Pb is similar to that of C−Cu, which shows that carbon has some solubility in the Pb solid solution at the growth temperature. It is reasonable to deduce that the solubility of carbon in nanometer-sized catalyst particles may be increased significantly; therefore, carbon atoms will be able to dissolve into the Pb nanoparticles and precipitate out to form graphitic structures according to the VLS mechanism.[17]

As stated above, the volatility of Pb is favorable for obtaining SWCNTs with no catalyst residues. However, the volatile character of Pb might also be adverse for the CVD growth of SWCNTs. In a gas flow of >900 °C under CVD conditions, it is important to maintain the metallic Pb particles before the nucleation and during the growth of the SWCNTs in the CVD process. We have designed several special methods for this purpose, including a low gas flow method, a fast-heating method, and a polyvinylpyrrolidone (PVP) assisted method. With these special methods, both random SWCNT networks and horizontally aligned SWCNT arrays on silicon wafers were prepared via Pb-catalyzed CVD. Both Raman spectroscopy and electrical transport measurement verified that the SWCNTs synthesized by using Pb as a catalysts are of high quality.

2. Experimental Section

2.1. CVD Growth of SWCNTs on Silicon Wafers by Using Pb As the Catalyst. In all experiments, silicon wafers with a silica layer on top was used as the growth substrates where the catalysts were loaded and the SWCNTs were grown. Methane was used as the feeding gas except where specified. A quartz tube (25 mm in diameter) equipped in a horizontal tube furnace with temperature controller was used as the CVD reactor. To avoid the release of poisonous Pb vapor into the air, a cold trap was installed into the exhaust of the CVD system.

2.1.1. Random SWCNT Networks Using the Low-Gas-Flow Method. The ethanol solution of $Pb(Ac)_2$ (1 mM) was dropped onto a piece of silicon wafer to act as the precursor of the Pb catalyst. The wafer was placed into a small interior quartz tube with the inner diameter of 5 mm and then mounted together into the quartz tube reactor.[18] The wafer was rapidly heated to 950 °C in Ar and then treated with 50 sccm of H_2 for 15 min. After that, the H_2 flow was decreased to 10 sccm, and then 5 sccm of CH_4 was also introduced into the system for 30 min. Finally, it was cooled under Ar flow to room temperature.

2.1.2. Random SWCNTs Grown Using the Fast-Heating Method. The ethanol solution of $Pb(Ac)_2$ (0.01 mM) was dropped onto the silicon wafer and then dried in air. Then the wafer with $Pb(Ac)_2$ catalyst was put into the quartz tube reactor and calcined in air at 700 °C for 5 min. After that, the wafer was pulled out of the center of the furnace. The furnace was continuously heated to 925 °C under an Ar atmosphere, and a flow of CH_4 (360 sccm) and H_2 (180 sccm) was then introduced into the system and the silicon wafer was quickly transferred to the center of the heated furnace. After 15 min of growth, the system was cooled to room temperature under Ar.

2.1.3. Random SWCNTs Grown with PVP-Capped Pb Precursor. The ethanol solution of $Pb(Ac)_2$ (0.5 mM) and PVP (250 mM counted by vinylpyrrolidone monomer) was dropped onto the silicon wafer and dried in air. Then the wafer was placed into the quartz tube reactor and calcined in air at 700 °C for 5 min. H_2 (300 sccm) was introduced into the system for 5 min, and within the next 10 min, the substrate was heated to and maintained at 900 °C under a H_2 flow. Subsequently, CH_4 (215 sccm) was introduced for the growth of SWCNTs for 15 min, followed by cooling the furnace in Ar to room temperature. SWCNTs were also grown on silica microspheres using this method for HRTEM imaging.

2.1.4. Random SWCNTs Grown with Ethanol As the Carbon Stock. The ethanol solution of $Pb(Ac)_2$ (0.01−1 mM) was dropped onto the silicon wafer and dried in air. Then the wafer with $Pb(Ac)_2$ catalyst was put into the quartz tube reactor and calcined in air at 700 °C for 5 min. The furnace was continuously heated to 900 °C under an Ar atmosphere, and then ethanol vapor was introduced into the furnace by bubbling H_2 through ethanol at a flow rate of 300 sccm. After 15 min of growth, the system was cooled to room temperature under Ar.

2.1.5. Horizontally Aligned Arrays of SWCNTs Grown Using the Fast-Heating Method. The same precursor solution with PVP for growing random SWCNTs was used. The catalyst precursor was patterned onto the silicon wafer by PDMS stamping. Horizontally aligned arrays of SWCNTs were then grown using the fast-heating method similar to that of the random SWCNTs' growth. The growth temperature was 950−970 °C; 360 or 320 sccm of CH_4 together with the same flow of H_2 was used for the CVD process.

2.1.6. Preparation of PbS Nanoparticles. Ten milliliters of 0.01 M $Pb(Ac)_2$ aqueous solution was added to 10 mL of 0.01 M Na_2S aqueous solution under stirring, and the solution was then stirred for 1 h. The black precipitates were washed with distilled water several times and then dispersed in ethanol.

2.1.7. Growth of SWCNTs with As-Prepared PbS Nanoparticles. The ethanol dispersion of the PbS nanoparticles from the previous procedure was dropped onto a silicon wafer or patterned by PDMS stamping. The growth methods of SWCNTs with PbS nanoparticles were nearly the same as that using $Pb(Ac)_2$ as the catalyst precursor with low feeding gas flow except that the reducing time was shortened to not longer than 10 min, because PbS is more volatile.

2.2. Characterization. Scanning electron microscopy (SEM, S4800, Hitachi, Japan, operated at 1 kV), high-resolution transmission electron microscopy (HRTEM, Hitachi 9000, operated at 100 kV), micro-Raman spectroscopy (Renishaw, Ramanscope-1000, with the excitation wavelength of 514.5 nm), and atomic force microscopy (AFM, SPI3800, operated at tapping-mode) were used to characterize as produced SWCNTs. Transmission electron microscopy (TEM, JEOL-200 CX) was used to characterize the as prepared PbS nanoparticles.

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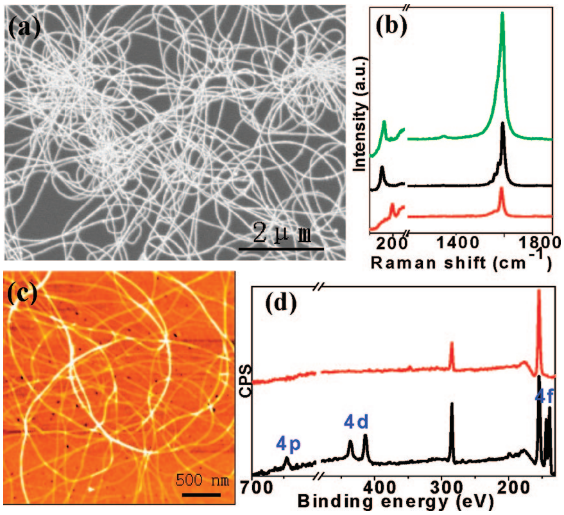

Figure 1. (a) SEM and (c) AFM images of SWCNTs grown at 950 °C with 1 mM $Pb(Ac)_2$ using low gas flow method. (b) Micro-Raman spectra of the corresponding sample. The RBM section was enlarged by 5 times. (d) XPS data of the silicon wafer with $Pb(Ac)_2$ catalyst before and after the growth of SWCNTs. Peaks marked with 4p, 4d, and 4f were electron characteristic peaks of Pb elements.

2.3. Electrical Transport Measurements. Highly p-doped Si wafers with a 500 nm SiO_2 dielectric layer were used as the substrates for the growth of ultralong SWCNTs and directly used as usual back gates for the transport measurements. Pd (40nm) was used as source/drain electrodes, which were patterned by E-beam lithography, E-beam evaporation, and standard lift-off processes. The electrical transport measurements of the back gated field effect transistors (FETs) based on single SWCNTs were carried out with a Keithley 4200 semiconductor characterization system.

3. Results and Discussion

3.1. Growth of Random SWCNT Networks with Pb As Catalyst. We have developed several methods to eliminate the excessive evaporation of Pb. The first is the low gas flow method.[18] The relative small space within the smaller quartz tube confined the volatilization of Pb, and the low gas flow reduced the amount of Pb vapor carried by the gas flow. Dense networks of random SWCNTs were prepared using this method with 1 mM of $Pb(Ac)_2$ as the catalyst precursor. Interestingly, as-prepared SWCNTs were often tangled with each other and formed flowerlike shapes (images a and c in Figure 1). Figure 1b shows the typical Raman spectra of the sample. The peaks of the radial breathing mode (RBM) at 153, 162, and 197 cm^{-1} correspond to SWCNTs with the diameters of 1.62, 1.53, and 1.26 nm, respectively (data are shown in Table S1 in the Supporting Information). The quite weak D-band shows that the SWCNTs prepared using this method were of high quality.

To clear up the possibility of the presence of Fe contamination before and during the CVD process and to ensure that it was Pb that acts as the catalyst, we undertook many careful investigations.[9] We used new quartz tubes for the Pb-catalyzed CVD experiments to avoid contamination; we repeatedly did experiments on blank samples without catalysts and did not find any trace of the formation of SWCNTs; and we carried out parallel experiments using other acetates (such as NaAc, $Ca(Ac)_2$ and $Zn(Ac)_2$) instead of $Pb(Ac)_2$ and found that still no carbon nanotubes were formed. Finally, we used X-ray photoelectron spectroscopy (XPS) to show that no Fe or other metal elements that normally can catalyze the SWCNTs' growth existed in our samples (Figure 1d). In the sample treated with H_2 but before CVD growth, the characteristic peaks of Pb including 646 eV (4p), 436 and 414 eV (4d), and 144 and 138 eV (4f) are clearly given. No catalytic metals other than Pb existed in our catalyst. And after the CVD process, no new element appeared. This result proves that it was Pb that catalyzed the growth of SWCNTs. What's more interesting, it was found that the characteristic peak of Pb disappeared after the growth of SWCNTs. This pointed out the absence of Pb catalyst in the as prepared SWCNT samples because Pb volatilizes during the process of growth. This provides us a strategy to obtain metallic residue-free SWCNT samples, which are very important for the study of intrinsic properties of SWCNTs.

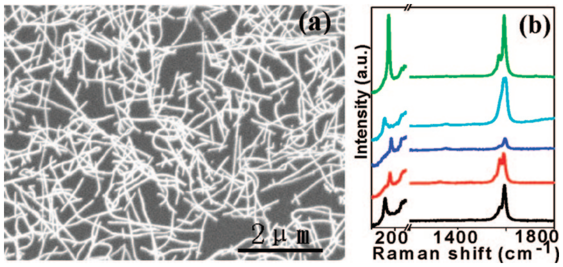

Figure 2. (a) SEM image of SWCNTs grown at 925 °C with 0.01 mM $Pb(Ac)_2$ catalyst using fast-heating method. The flow velocity of CH_4/H_2 were 360/180 sccm. (b) Micro-Raman spectra of the corresponding sample. The RBM section was enlarged by 5 times.

The second method we used was fast-heating, which was usually employed to prepare ultralong SWCNTs.[19] But the flow rate we used here was lower than that for ultralong SWCNTs. Because the silicon wafer with the catalyst was transferred into the center of the heated furnace just before the beginning of the growth process, the volatilization of Pb would be reduced enormously. Therefore, a dense random SWCNT network on silicon wafer was prepared (Figure 2a). RBM bands at 157, 159, 173, 180, and 185 cm^{-1} were found in the Raman spectra (Figure 2b) and the corresponding diameters of the SWCNTs were 1.58, 1.56, 1.43, 1.38, and 1.34 nm, respectively (data shown in Table S1 of the Supporting Information). The intensity of the D-band was also quite weak.

The third method of reducing the volatilization of Pb catalyst was to add a polymer into the catalyst precursor solution. An appropriate amount of PVP was added to the $Pb(Ac)_2$ solution to obtain a solution of $Pb(Ac)_2$/PVP with the molar ratio of 1/50. Due to the protection of PVP, the catalyst precursors can be maintained on the silicon wafer longer. It was found that when PVP was used, uniformly distributed SWCNTs with enhanced density were obtained on the silicon wafer using the normal heating method (images a and c in Figure 3). The inset of Figure 3c shows the HRTEM image of a SWCNT with the diameter of 2 nm

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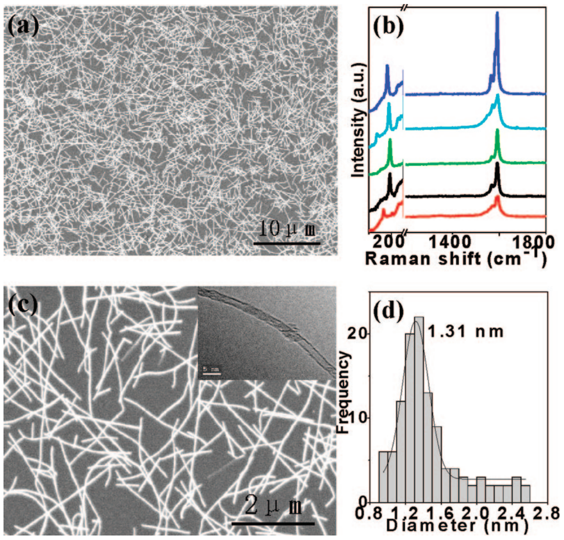

Figure 3. (a, c) SEM images of SWCNTs grown on silicon wafer with $Pb(Ac)_2$/PVP (mol. 1/50) catalyst using normal heating method. The HRTEM image of a SWCNT grown on silica microsphere was shown in the inset of c. (b) Micro-Raman spectra of the corresponding sample. The RBM section was enlarged by 5 times for clarity. (d) Size distribution analysis from 115 SWCNTs measured in AFM images. Gaussian fit was done and given the mean diameter of 1.44 nm.

grown under the same conditions on silica spheres.[20] The diameters of the SWCNTs were determined by measuring the heights in AFM images. The statistical chart of Figure 3d shows the size distribution of the obtained SWCNTs. A Gaussian fit was done and gave the most probable diameter of 1.31 nm and mean diameter of 1.44 ± 0.37 nm. RBM bands at 136, 165, 181, 188, and 194 cm^{-1} in the Raman spectra (Figure 3b) shows the presence of SWCNTs with diameters of 1.82, 1.5, 1.37, 1.32, and 1.28 nm, respectively (data shown in Table S1 of the Supporting Information), which was consistent with the result from the AFM images. The quite weak intensity of the D-band shows again the high quality of the obtained SWCNTs.

Ethanol can also be the carbon stock instead of methane for the growth of SWCNTs with $Pb(Ac)_2$ catalyst. As the pyrolysis speed of ethanol is much faster than methane, dense networks of random SWCNTs can be prepared with the volatile Pb catalyst. In the process, ethanol was introduced into the system by the H_2 flow (300 sccm). The SWCNTs as prepared contained two distinct extended forms, that is, aligned SWCNTs grown along the direction of the gas flow, and random SWCNTs without uniform growth direction (Figure S1 in the Supporting Information). And the density of the SWCNTs was larger when 1 mM $Pb(Ac)_2$ was used compared with 0.1 mM.

3.2. Preparation of Horizontally Aligned Ultralong SWCNT Arrays with Pb As Catalyst. Horizontally aligned arrays of SWCNTs were prepared with $Pb(Ac)_2$/PVP (mol. 1/50) catalyst using fast-heating method. Large-area and well-aligned SWCNT arrays were obtained (Figure 4). The markers on silicon wafer (Figure 4c) enabled us to carry out the AFM and Raman measurements on the same SWCNT. Figure 4d showed the AFM image of one SWCNT in Figure 4c. The corresponding Raman spectrum (Figure 4e) gave a RBM band at 173 cm^{-1} and the corresponding diameter of this SWCNT was 1.43 nm.

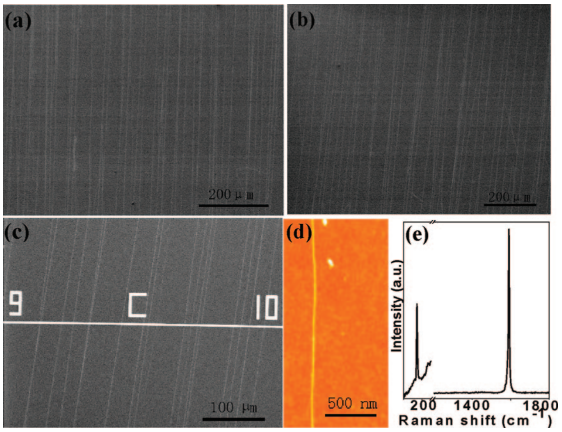

Figure 4. (a−c) SEM images of horizontally aligned arrays of SWCNTs grown at 970 °C with $Pb(Ac)_2$/PVP (mol. 1/50) catalyst using the fast-heating method. The flow velocity of CH_4/H_2 was 360/360 sccm. (d) AFM image and (e) the corresponding Raman spectrum of a SWCNT in sample c.

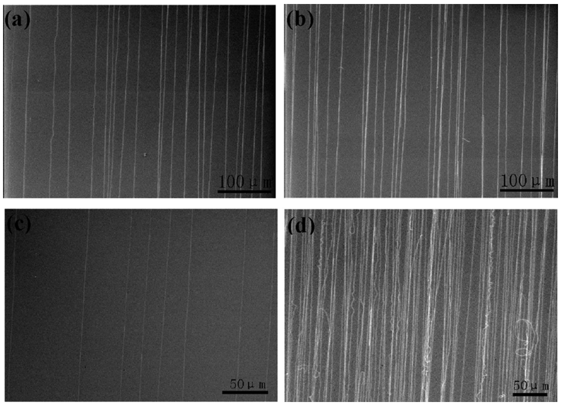

Figure 5. SEM images of horizontally aligned arrays of SWCNTs grown with $Pb(Ac)_2$/PVP (mol. 1/50) catalyst using fast-heating method. (a, b) Grown at 950 and 970 °C, respectively, with the same flow rate of CH_4/H_2 (360/360 sccm). (c, d) Grown at 950 °C, and the flow rates of CH_4/H_2 were (c) 360/450 sccm and (d) 320/320 sccm, respectively.

The suitable growth temperature for horizontally aligned SWCNT arrays was 950−970 °C (images a and b in Figure 5). Lower or higher growth temperatures would lead to SWCNT arrays with lower density or poor quality. The density of the SWCNTs array was mainly affected by the flow rate of CH_4 and H_2 and the proportion between CH_4 and H_2. Lowering the total gas flow and reducing the proportion of H_2 used will benefit higher density as shown in Figure 5. The optimal proportion between CH_4 and H_2 was 1/1. For example, when the flowrate of CH_4 was fixed to 360 sccm, the densities of the SWCNT arrays as prepared were 2 SWCNTs (Figure 5c) and 5 SWCNTs (Figure 5a) in 100 μm for the H_2 flowrate of 450 and 360 sccm, respectively. A SWCNT array with higher density (20 SWCNTs in 100 μm) was obtained with a lower gas flow of 320 sccm CH_4 and 320 sccm H_2 (Figure 5d).

3.3. Growth of SWCNTs with PbS Nanoparticles As the Catalyst Precursors. Besides the $Pb(Ac)_2$ catalyst, PbS nanoparticles can also be used as the catalyst precursor for the growth of SWCNTs. The PbS nanoparticles used had a

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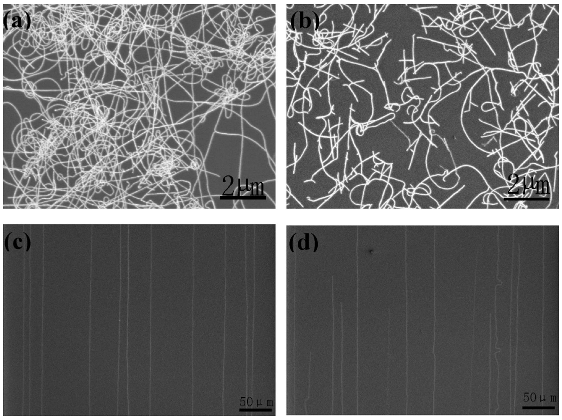

Figure 6. SEM images of the SWCNTs grown with PbS nanoparticles as catalyst precursors. (a) SWCNTs grown at 950 °C using the low-gas-flow method; the H_2 treatment lasted for 5 min. (b) SWCNTs grown at 925 °C using the fast-heating method; the flow rate of CH_4/H_2 was 360/180 sccm. (c, d) Horizontally aligned arrays of SWCNTs with low-density grown at 950 °C using the fast-heating method; and the flow rate of CH_4/H_2 was 360/360 sccm.

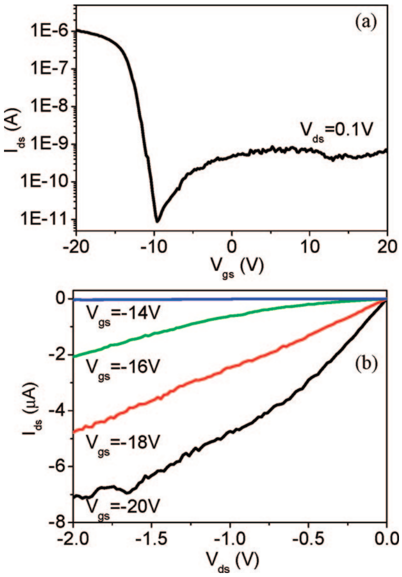

Figure 7. (a) $I_{ds}-V_{gs}$ and (b) $I_{ds}-V_{ds}$ curves of the single tube device. Channel length is 2 μm.

diameter of ~10 nm (see Figure S2 in the Supporting Information). The growth methods of SWCNTs with PbS nanoparticles as catalyst precursors were nearly the same as that with the $Pb(Ac)_2$ catalyst. Similar random SWCNT networks were prepared using both the low gas flow method and the fast heating method (images a and b in Figure 6). The amount of SWCNTs was less because of the higher volatility of PbS nanoparticles compared with $Pb(Ac)_2$. Horizontally aligned arrays of SWCNTs with low density were prepared at 950 °C using the fast-heating method (images c and d in Figure 6). Because of the higher volatility of PbS, it may evaporate easily and recondense to form new nanoparticles at different locations. Therefore, we can see the ends of the ultralong SWCNTs where they started to grow from the middle of the silicon wafer in Figure 6d.

3.4. Transport Measurement of FETs Fabricated with Individual SWCNTs from the Pb Catalyst. To further investigate the quality of the SWCNTs prepared with the Pb catalyst, we carried out electrical transport measurements for single tubes in the horizontally aligned ultralong arrays (as shown in Figure 4) with the highly doped silicon wafer substrates as the gates and Pd as the source/drain (S/D) electrodes. It was found that the transport behavior of the SWCNTs from Pb is very similar to those from Fe.[21] In Figure 7, we show the typical electrical characteristics of a back gated p-type SWCNT based FET with 2 μm channel length. Low bias $I_{ds}-V_{ds}$ characteristics of the on state ($V_{gs} = -20$ V) (Figure 7b) were perfectly linear, indicating SWCNTs and S/D electrodes were ohmic-contacted. This device had an on/off ratio of 1×10^5 (Figure 7a), and the on state current could reach the microamp scale at room temperature. The result of the electrical transport measurement demonstrated that our devices had high performance and further verified our SWCNTs synthesized by using Pb as catalysts were of high quality.

Conclusions

In summary, we found that Pb can be a good catalyst for CVD growth of SWCNTs. However, it is necessary to avoid the excessive volatilization of Pb during the CVD process. Three methods were employed for this purpose, that is, a low gas flow method, a fast-heating method, and a PVP assisted method. Dense random SWCNT networks and density-controlled horizontally aligned ultralong SWCNT arrays were prepared with $Pb(Ac)_2$ and PbS nanoparticles as catalyst precursors. Moreover, both CH_4 and ethanol can be used as carbon stocks. The single SWCNT FET devices showed high performance in electrical transport measurements.

Relying on the volatility of Pb, we realized direct growth of SWCNTs without metallic impurities on the substrates. SWCNTs prepared using the Pb catalyst were of high quality and can be used for the study of intrinsic properties of SWCNTs, for biological applications, and in building nanodevices because of the absence of residual metallic catalysts.

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Supporting Information Available: Data of the RBM frequency and corresponding diameters of the SWCNTs in Raman spectra (PDF). This material is available free of charge via the Internet at http://pubs.acs.org.

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