THE JOURNAL OF PHYSICAL CHEMISTRY B LETTERS

2005, *109,* 11420−11423
Published on Web 05/18/2005

Controllable Interconnection of Single-Walled Carbon Nanotubes under AC Electric Field

Zhuo Chen, Yanlian Yang,† Fang Chen, Quan Qing, Zhongyun Wu, and Zhongfan Liu*

Center for Nanoscale Science and Technology (CNST), College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, P. R. China

Received: April 9, 2005; In Final Form: May 8, 2005

We demonstrated the controllable interconnection of single-walled carbon nanotubes (SWNTs) under alternating current (ac) electric field. The interconnected carbon nanotubes were found to be parallel with the electric flux and increased abruptly with deposition time following a self-accelerating process. Theoretical simulation indicates that the alignment and the interconnection of carbon nanotubes were induced by the dielectrophoresis force and the electric field redistribution at the nanotube apexes.

Single-walled carbon nanotubes (SWNTs) have attracted wide attention for their unique physical and chemical properties in recent years. Many works have focused on nanodevices based on individual SWNTs or bundles, such as transistors,[1−3] diodes,[4,5] superconductor quantum wires,[6] and sensors,[7,8] while for the practical applications of SWNTs, especially for the integration of nanotube-based devices, the controllable alignment and interconnection of SWNTs are necessary. Many efforts have been made up to now along this direction. Liu et al.[9] reported that Au nanoparticles could be used as the joint to connect thiol-derivatized carbon nanotubes. Ajayan's group reported that the crossing SWNTs could be joined by electron beam welding to form molecular junctions.[10] Recently, electric field has been frequently utilized to manipulate the nanoscale materials (such as proteins,[11] nanoparticles,[12] and carbon nanotubes,[13−17] etc.) into well-aligned structures. For instance, Velev's group reported that microwires could be assembled from suspensions of metallic nanoparticles by applying electric field, and the interconnected nanoparticles showed good electrical conductivity.[12] Similar interconnection behavior is expected with the SWNTs under electric field, which has been demonstrated in this work.

SWNTs were produced by the chemical vapor deposition (CVD) method using Fe catalyst supported by MgO.[18] SWNTs (ca. 500 nm − 2 μm long) were dispersed in dimethylformamide (DMF) under ultrasonication. Au electrodes of 100-nm thickness with 10-nm-thick Ti adhesion layer were fabricated by conventional UV lithography and lift-off technique on Si/SiO_2 substrates with a 5-μm gap. The strategy of alignment and interconnection of carbon nanotubes under the ac electric field is illustrated in Figure 1a. The gold electrodes were first modified by octadecylthiol (ODT) self-assembled monolayer (SAM). The ODT monolayer was found to effectively reduce the contamination of carbonaceous impurities involved in SWNT solution, possibly owing to the introduced hydrophobic surface effect. An ac electric voltage with a magnitude of 1.25 V peak to peak and a frequency of 5 MHz was then applied to the electrodes for a certain time after immersion into the SWNT suspension. Finally, the electrodes were rinsed thoroughly with DMF and dried with N_2 gas. All the experiments were conducted at room temperature.

Figure 1b−d showed the scanning electron microscope (SEM) images of the aligned and interconnected SWNTs between two electrodes. After application of an ac electric field (2.5×10^5 V/m) for 20 min, alignment of SWNTs (Figure 1b) was observed at electrode surfaces with their long axes parallel to the electric flux.[14] More importantly, interconnection of individual SWNTs or bundles occurred along the electric field direction as seen in Figure 1c,d. In Figure 1c, two SWNT bundles were connected together at their ends and protruded out from one electrode. Such an interconnection can be extending to bridge the two electrodes as shown in Figure 1d.

The driving force of the interconnection of SWNTs under ac electric field is believed to be the high electric field gradient generated between two electrodes. Velev et al.[12] have reported that gold nanoparticles can be interconnected into microwires along the direction of the field gradient under external ac electric field by a collective effect. A similar physical process is expected for the present SWNT system. A dipole moment can be easily induced for the large π-conjugated SWNTs under an electric field. Such polarized nanotubes would undergo a directional movement along the electric field gradient under the inhomogeneous electric field, which is commonly referred to as a dielectrophoresis (DEP) effect. The DEP force can be expressed as[13]

$$\vec{F}_{\mathrm{DEP}} \propto \epsilon_{\mathrm{m}} \frac{\epsilon_{\mathrm{p}} - \epsilon_m}{\epsilon_{\mathrm{p}} + 2\epsilon_{\mathrm{m}}} \nabla {E_{\mathrm{rms}}}^2 \qquad (1)$$

where ϵ_p and ϵ_m are the dielectric constants of carbon nanotubes and solvent medium, respectively. E_{rms} is the average field strength. When an electric field was applied across two electrodes, deposition of SWNTs on the electrode surface occurred, which led to a remarkable change of electric field distribution. The electric field distribution in the existence of SWNTs was simulated using the *Matlab* program (MathWorks, Inc.) on the actual device geometry shown in Figure 1a, supposing that the SWNT has a radius of 0.5 nm and a length of 1 μm. The simulation result is given in Figure 2a,b.

* To whom correspondence should be addressed. Phone and fax: 86-10-6275-7157. E-mail: zfliu@pku.edu.cn.
† Current address: National Center for Nanoscience &Technology, Beijing, 100080, P. R. China.

10.1021/jp051848i CCC: $30.25

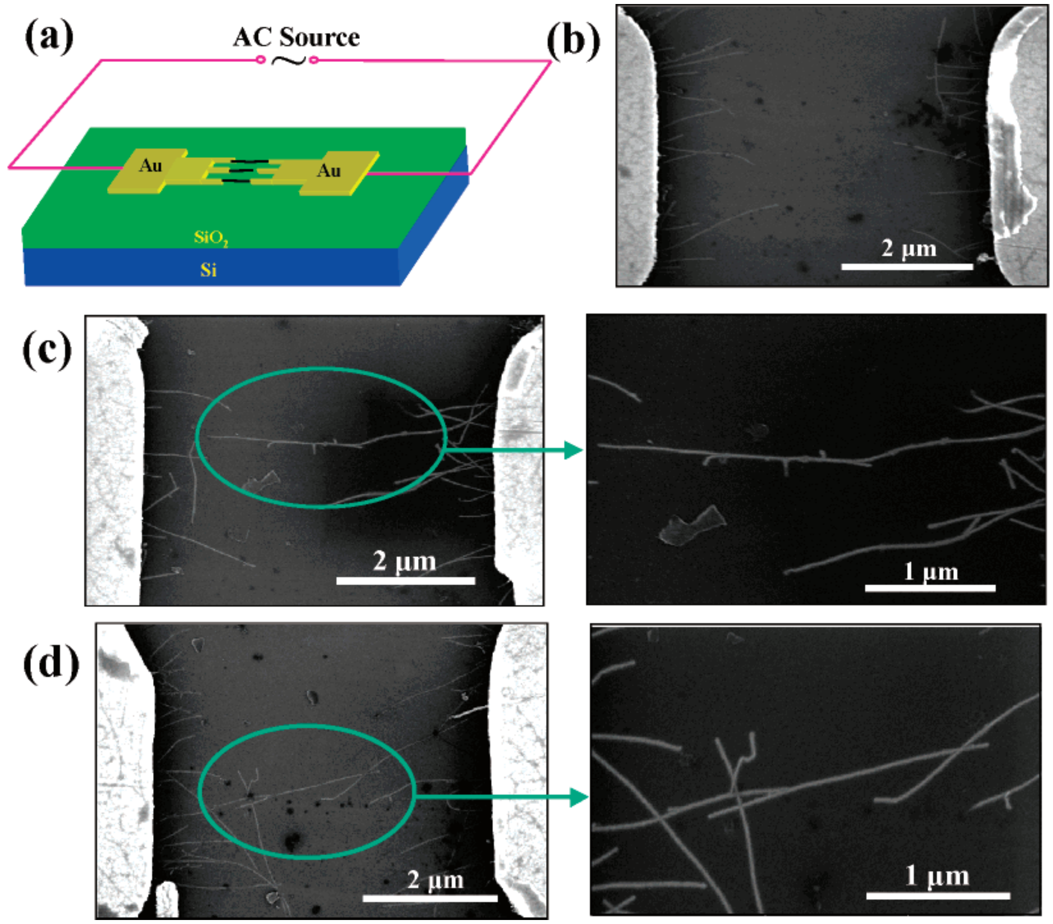

Figure 1. (a) Schematic illustration of the experimental system for the alignment and interconnection of SWNTs under ac electric field (2.5×10^5 V/m, 5 MHz). (b) Alignment of SWNTs on the electrode surfaces with their long axes along the electric field flux. (c) Interconnection of two SWNT bundles that protruded from one electrode. (d) Interconnection of three SWNT bundles one by one which bridged two electrodes. The right-hand figures in (c) and (d) are the closeups of the left-hand ones.

Obviously, the nanotube apex demonstrated extremely high strength and gradient of electric field (see the deep dark region of Figure 2b). This indicates that the DEP force acting on the solution-phase carbon nanotubes gets greatly enhanced at the surface-fixed nanotube apex, taking account of eq 1, resulting in the directional movement of solution-phase carbon nanotubes and connection to the surface-fixed nanotube apexes along the field gradient. Similar processes could be repeated until two electrodes are completely bridged by the interconnected carbon nanotubes.

Figure 2c,d showed the experimental results supporting the above theoretical interpretation. The electrode was first immersed into the SWNT solution for 15 min under an ac electric field (2.5×10^5 V/m, 5 MHz). A number of SWNTs were deposited on the electrode surfaces as seen in Figure 2c. Then, 15-min-longer deposition of SWNTs from DMF solution was performed under the same experimental conditions. As shown in Figure 2d, a single nanotube bundle was just deposited between the two protruding nanotubes, forming a bridge between two electrodes. This strongly suggests that the carbon nanotubes preferred to move toward the higher field gradient region created in the predeposited nanotube apexes.

It seems that the interconnection of SWNTs under ac electric field is an accelerated process. Figure 3a–d showed the time dependence of interconnencted nanotubes between two electrodes under electric field. Very few nanotubes were observed with 10 min of ac deposition, as seen in Figure 3a. Interconnection of carbon nanotubes occurred at 20 min of ac deposition (Figure 3b) and increased remarkably at 25 min of ac deposition (Figure 3c). Huge numbers of carbon nanotubes have been deposited between two electrodes at 30 min of ac deposition, forming a nanotube film as shown in Figure 3d. The alignment structure of carbon nanotubes was most likely reflecting the distribution of electric field between two electrodes.

The electrical conductivity between two electrodes was followed at different ac deposition times by a Keithley 4200 measurement system (U.S.A.). The result is shown in Figure 3e. When the deposition time was less than 20 min, the electrical conductivity was extremely low, because the nanotube bridges have not been created between two electrodes as seen from the SEM image (e.g., Figure 3a). Together with the increase of deposition time, electrical connection between two electrodes with carbon nanotubes occurred, and the conductivity was increased to 5.0×10^{-7} S (20 min), 2.7×10^{-5} S (25 min), and 1.1×10^{-3} S (30 min). The nanotube bridges estimated from SEM images were approximately 10, 100, and several thousands, respectively. An abrupt increase of conductivity was observed after 25 min deposition, strongly suggesting the occurrence of accelerated interconnection of carbon nanotubes.

As mentioned above, when a carbon nanotube was attached to the electrode surface, the electric field would be redistributed. The field strength around the nanotube apex should be much higher, as indicated in Figure 2b. The enhancement factor γ at the apex region of carbon nanotube can be expressed as[19]

$$\gamma = 1.0782(l/\alpha + 4.7)^{0.9152} \qquad (2)$$

where l and a are the length and radius of the carbon nanotube,

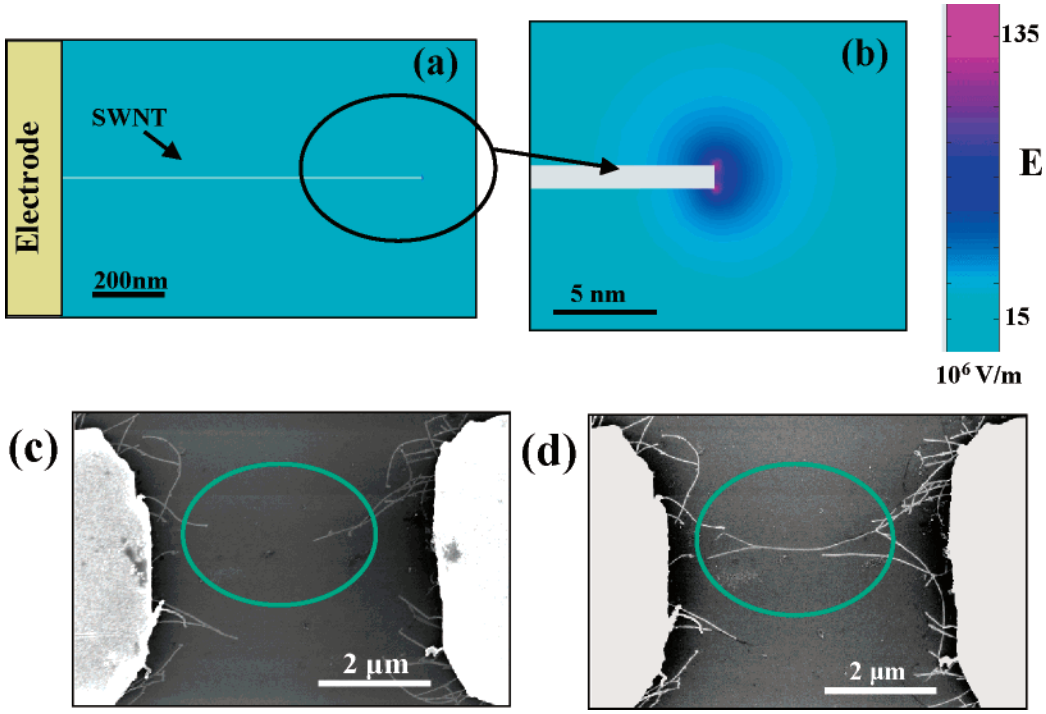

Figure 2. (a) Electric field distribution after one SWNT was predeposited on the electrode simulated using *Matlab* program. (b) Clsoeup of (a). (c) SEM image of SWNTs deposited on the electrode surfaces under ac electric field for 15 min. (d) Deposition for 15 min more after (c).

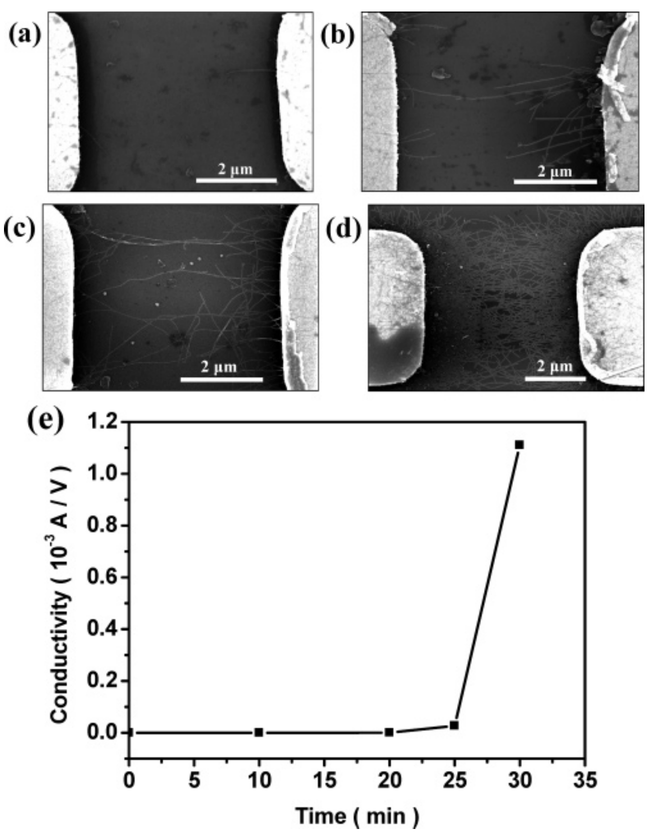

Figure 3. SEM images of SWNTs deposited between two electrodes under ac electric field for 10 min (a), 20 min (b), 25 min (c), and 30 min (d). (e) Dependence of electrical conductivity measured ex situ between two electrodes with deposition time under ac electric field.

respectively. For a 1-μm-long and 0.5-nm-radius nanotube, the enhancement factor would be around 1100. Therefore, such a redistribution of electric field at the nanotube apex creates a local field with high intensity and gradient. The interconnected nanotubes can be functioning as a longer tube, which further increases the enhancement factor γ, as deduced from eq 2. As a result, the DEP force becomes even larger, which accelerates the interconnection process.

With the above experimental evidence and theoretical consideration, we can conclude that the dielectrophoresis force and the electric field redistribution at the carbon nanotube apexes are responsible for the interconnection phenomenon. Our results demonstrate that SWNTs can be aligned and interconnected between two electrodes under ac electric field. The aligning and interconnecting process can be easily controlled by deposition time. We believe that such a controlled interconnection of SWNTs may provide a possible way to fabricate carbon nanotube-based devices. A direct wiring to the external electrodes is also a very important advantage for the device evaluation. The strategy used in this work may also be extended to other nanotube and nanowire systems.

Acknowledgment. This work was supported by the National Natural Science Foundation of China (NSFC, 60301001, 90301006) and the Ministry of Science and Technology (MOST, 2001CB6105).

References and Notes

(1) Martel, R.; Schmidt, T.; Shea, H. R.; Hertel, T.; Avouris, Ph. *Appl. Phys. Lett.* **1998**, *73*, 2447.
(2) Heinze, S.; Tersoff, J.; Martel, R.; Derycke, V.; Appenzeller, J.; Avouris, Ph. *Phys. Rev. Lett.* **2002**, *89*, 106801.
(3) Javey, A.; Guo, J.; Wang, Q.; Lundstrom, M.; Dai, H. J. *Nature (London)* **2003**, *424*, 654.
(4) Lee, K.; Wu, Z.; Chen, Z.; Ren, F.; Pearton, S. J.; Rinzler, A. G. *Nano Lett.* **2004**, *4*, 911.
(5) Lee, J. U.; Gipp, P. P.; Heller, C. M. *Appl. Phys. Lett.* **2004**, *85*, 145.
(6) Wei, Y. D.; Wang, J.; Guo, H.; Mehrez, H.; Roland, C. *Phys. Rev. B* **2001**, *63*, Art. 195412.
(7) Chopra, S.; Pham, A.; Gaillard, J.; Parker, A.; Rao, A. M. *Appl. Phys. Lett.* **2002**, *80*, 4632.
(8) Chen, R. J.; Choi, H. C.; Bangsaruntip, S.; Yenilmez, E.; Tang, X. W.; Wang, Q.; Chang, Y. L.; Dai, H. J. *J. Am. Chem. Soc.* **2004**, *126*, 1563.

(9) Liu, J.; Rinzler, A. G.; Dai, H. J.; Hafner, J. H.; Bradley, R. K.; Boul, P. J.; Lu, A.; Iverson, T.; Shelimov, K.; Huffman, C. B.; Rodriguez-Macias, F.; Shon, Y. S.; Lee, T. R.; Colbert, D. T.; Smalley, R. E. *Science* **1998**, *280*, 1253.

(10) Terrones, M.; Banhart, F.; Grobert, N.; Charlier, J. C.; Terrones, H.; Ajayan, P. M. *Phys. Rev. Lett.* **2002**, *89*,Art. 075505.

(11) Hughes, M. P.; Morgan, H. *Anal. Chem.* **1999**, *71*, 3441.

(12) Hermanson, K. D.; Lumsdon, S. O.; Williams, J. P.; Kaler, E. W.; Velev, O. D. *Science* **2001**, *294*, 1082.

(13) Krupke, R.; Hennrich, F.; von Lohneysen, H.; Kappes, M. M. *Science* **2003**, *301*, 344.

(14) Chen, X. Q.; Saito, T.; Yamada, H. *Appl. Phys. Lett.* **2001**, *78*, 3714.

(15) Nagahara, L. A.; Amlani, I.; Lewenstein, J.; Tsui, R. K. *Appl. Phys. Lett.* **2002**, *80*, 3826.

(16) Diehl, M. R.; Yaliraki, S. N.; Beckman, R. A.; Barahona, M.; Heath, J. R. *Angew. Chem., Int. Ed.* **2002**, *41*, 353.

(17) Chen, Z.; Yang, Y. Y.; Wu, Z. Y.; Luo, G.; Xie, L. M.; Liu, Z. F.; Ma, S. J.; Guo, W. L. *J. Phys. Chem. B* **2005**, *109*, 5473.

(18) Liu, Z. F.; Shen, Z. Y.; Zhu, T.; Hou, S. F.; Ying, L. Z.; Shi, Z. J.; Gu, Z. N. *Langmuir* **2000**, *16*, 3569.

(19) Read, F. H.; Bowring, N. J. *Nucl. Instrum. Methods, A* **2004**, *519*, 305.