

Dielectrophoretic Deposition of Carbon Nanotubes with Controllable Density and Alignment

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ABSTRACT

Controlled deposition of carbon nanotubes (CNTs) across desired electrodes is important for the fabrication of nanoelectronic devices. Dielectrophoresis (DEP) has been recognized as a convenient and affordable technique for the deposition of nanotubes and nanowires on electrodes. Although DEP has been quite well studied for dielectric particles, the application for depositing nanotubes is still at the early stage of development. Here, we show that multi-walled CNTs can be deposited by DEP with controllable density and degree of alignment.

INTRODUCTION

Carbon nanotubes are a popular nanomaterial due to their unique structural and electronic properties. These characteristics have facilitated their use in a wide array of devices and applications such as field effect transistors [1-5], chemical and biological sensors [6,7], scanning probe microscopy tips [8 - 10] and field emitters [11, 12]. However, before we can begin to take advantage of the desirable the properties of CNTs, they must be placed in the proper position for each application.

Techniques do exist for placement of CNTs, but each comes with its own associated drawbacks. Spin coating of nanotubes, for example, is cheap and simple, but lacks control of position and distribution [5, 13]. Direct growth requires high temperatures and suffers from poor selectivity and non-compatibility [14, 15], while manual attachment is unviable for devices of any significant density due to the enormous amount of time necessary to place individual CNTs.

A technique does exist that is quick, cheap and results in well-aligned tubes. This technique, called dielectrophoresis (DEP), involves alignment of solution-dispersed CNTs aligning between electrodes by application of an applied AC electric field. Although DEP has been quite well studied for dielectric particles, the application for depositing nanotubes and nanowires are still at the early stage of development. We have demonstrated that DEP can be used and that the density and alignment of CNTs in the electrode gap is controllable by varying the AC field's strength and frequency.

EXPERIMENT

Multi-walled CNTs were grown in-house by a dual plasma-enhanced chemical vapor deposition (PECVD) process [16]. This yields CNT directly on a substrate with lengths and widths of roughly 2 μm and 50 nm, respectively. After growth, the 7 mm diameter growth region had its CNTs removed and placed in ethanol for dispersion to a density of roughly 1×10^8 CNTs per mL. This suspension was placed in an ultrasonicating bath for 2 hours in order to disperse the CNTs without surfactant. Electrodes were patterned on glass substrates by pulsed laser deposition of an iron film to a thickness of 50 nm.

Before deposition, the electrodes were connected to a WAVETEK Sweep Generator model 22 and the AC field was turned on. Next, a 40 μL drop of the ethanol/CNT dispersion was applied directly to the gap giving it time to completely dry before the applied field is turned off. The results were studied using a Hitachi S4700 field-emission scanning electron microscope (SEM).

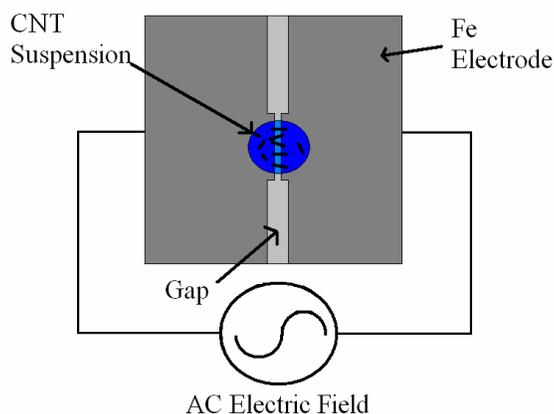


Figure 1. The DEP experimental setup. An AC electric field is applied between Fe electrodes before a CNT suspension is dropped into the gap. The field is left on until the drop is completely dry.

DISCUSSION

First we find that the AC field is responsible for alignment of the CNTs. This was done by comparison of tubes in the gap deposited without and with a 10 V peak to peak (V_{pp}), 5 MHz electric field (E). Figure 2 shows the difference: without field the tubes are randomly placed on and between the electrodes. These CNTs are well oriented in the direction of the field and attached to the gap when an electric field is applied. In addition, CNTs are capable of forming chains by attaching tail-to-tip once one tube has contacted the electrode.

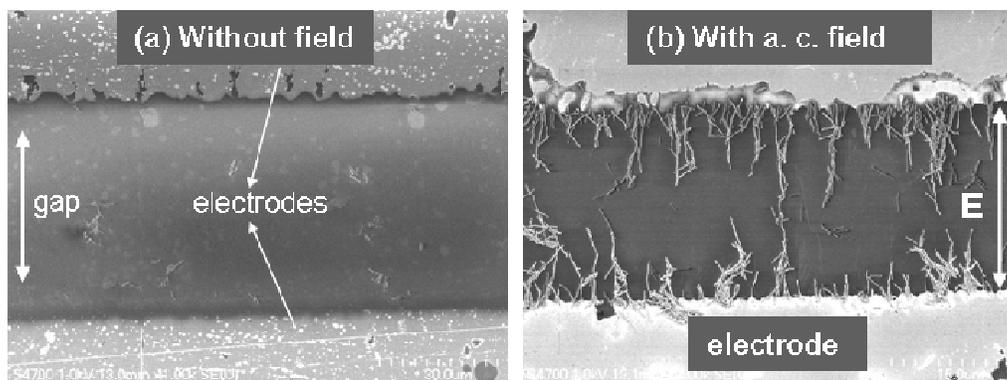


Figure 2. SEM showing the effects of AC fields on orientation of CNTs. (a) Tubes deposited with no field. (b) Tubes deposited with a $V = 10 V_{pp}$ and 5 MHz AC electric field. Gap distance $\sim 25 \mu\text{m}$.

In order to observe the effect of applied field strength, the frequency was fixed to 1 MHz while varying the voltage from 12 - 20 V_{pp}; Figure 3 shows the results of this test. Clearly the density (number of CNTs per unit length of the electrode gap) of tubes increases along with increasing field strength. Alignment is generally good with no apparent dependence on the field strength. This is, therefore, one method of controlling the spacing between CNTs, which may be important for fabrication of density-controlled parallel nanoelectronic devices.

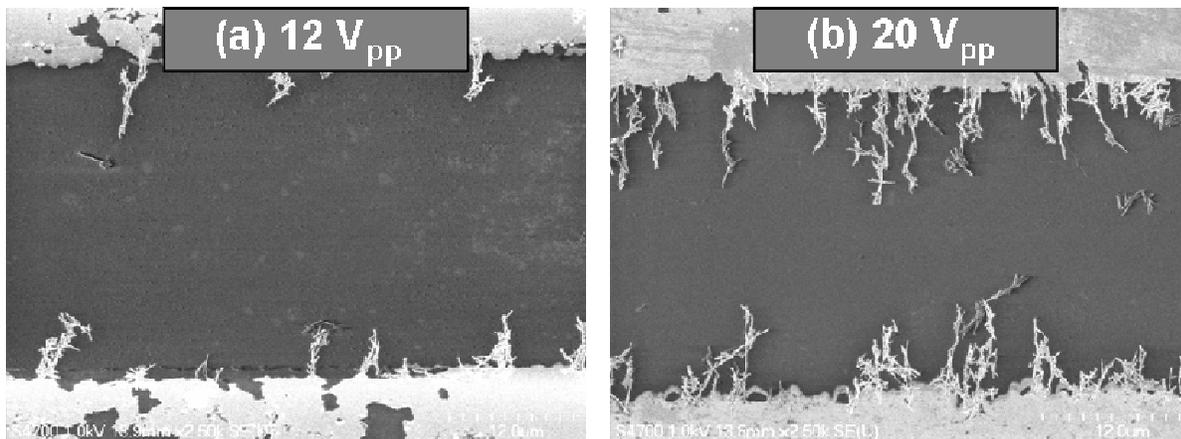


Figure 3. SEM images of PECVD grown CNTs subjected to AC fields with 1 MHz frequencies, but varying field strengths: (a) $V = 12V_{pp}$, (b) $V = 20V_{pp}$. Gap distance $\sim 25 \mu\text{m}$.

Next, the voltage was held at 20 V_{pp} and the frequency varied in order to study the effects of AC frequency on DEP; the results are shown in Figure 4. Low frequencies (500 Hz) showed the CNTs clumping on the gap with little to no alignment. Increasing the field to 1 MHz showed increased alignment; most tube bundles show better alignment than the clumps seen at 50 Hz. At 5 MHz the CNTs show alignment, but a remarkably lower density; the bundles of tubes are farther apart and contain fewer tubes per bundle. This shows that the degree of alignment and CNT density can be tuned by controlling the AC frequency of the applied field.

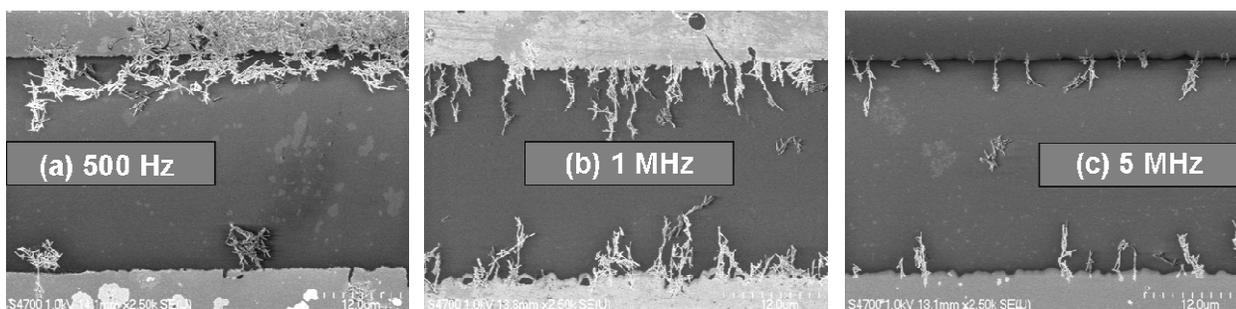


Figure 4. SEM images of PECVD grown CNTs subjected to an applied AC field of $V = 20 V_{pp}$ at varying frequencies: (a) $f = 500 \text{ Hz}$, (b) 1 MHz, (c) 5 MHz. Gap distance $\sim 25 \mu\text{m}$.

All results were reproduced and confirmed by DEP with CNTs grown by thermal CVD processes as well. For example, the tendency of the CNT density to decrease once the AC frequency has passed a critical frequency is shown in Figure 5. Here a 20 V_{pp} field was used and clearly shows better alignment and lower density at 5 MHz than 3 MHz.

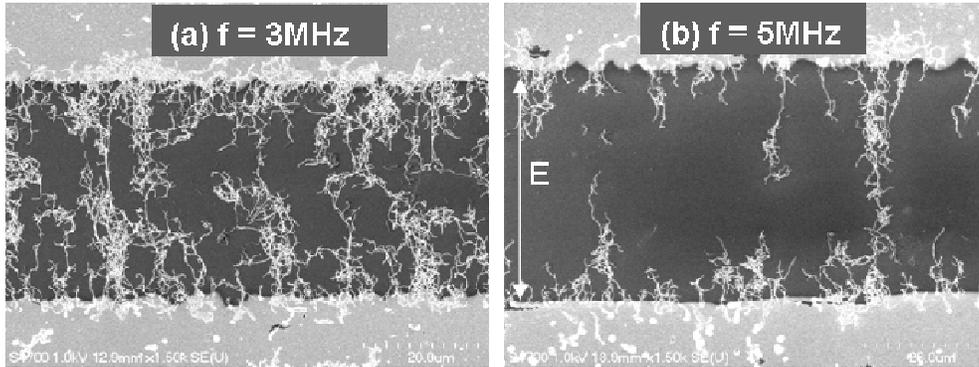


Figure 5. Thermal CVD grown CNTs, dispersed with 20 V_{pp} applied. Increasing the frequency of the AC field shows a reduction in tube density from 3 MHz (a) to 5 MHz (b). Gap distance ~ 30 μm.

Mechanisms

The alignment of CNTs across the gap is due to the interaction between the field-induced dipole moments of the CNTs and the applied electric field. Inside the field the CNTs become polarized and possess a dipole moment given by

$$P = LQ \quad (1)$$

where L is the length of the nanotube and Q is the charge of polarization at each end. Because of this polarization the field exerts a torque on the CNTs given by

$$\tau = P \times E = PE \sin\theta = LQE \sin\theta \quad (2)$$

where E is the applied electric field and θ is the angle between the axis of the CNT and the direction of E . The torque causes the CNTs to rotate and align in the direction of the applied field. Inhomogeneous fields bring about nanotube migration toward the regions where the field is the highest: the electrodes. Once a tube touches an electrode, it acquires the potential of the electrode and can attract CNTs to itself, leading to the chains observed by the SEM. Since higher electric fields result in higher field-induced forces on the CNTs, higher electric field yield a higher density of oriented tubes.

It is much harder to determine the role of frequency. We have found, however, that rather than directly taking part in the alignment process, the AC frequency helps to keep the nanotubes dispersed, with the level of dispersion proportional to the frequency. In suspension, the CNTs want to stick together and make clumps due to van der Waals forces between them, a process that begins immediately after sonication. CNTs that are better dispersed are easier to

align, so it is clear from the results shown in Figure 4 that the degree of alignment and, therefore, dispersion increases with AC frequency. Further, it is apparent that lower frequencies cannot overcome the attractive forces which result in the extreme clumping and poor alignment of Figure 4(a), but it is not clear why the density decreases at higher frequencies. We did notice that strength and frequency of the applied field required to orient CNTs of different morphologies are different. Thermal CVD grown CNTs [17, 18] are longer and thinner (13 μm x 20-30 nm) and more entangled than the PECVD tubes and are grown tangled, while PECVD tubes are separate. Frequencies higher than those used for the PECVD tubes were needed to orient the thermally grown CNTs, further elucidating the role of frequency in aided dispersion.

The increase of CNT deposition density at higher magnitude of applied voltage described here is consistent with other reports [19, 20]. However, Seo *et al.* shown that the degree of alignment of the deposited single wall CNTs is degraded after an optimum applied voltage [19]. The increase of CNT deposition density at a higher applied frequency is also consistent with the reported observation [20]. We found that an optimum frequency value occurred after which the deposited density decreases again.

CONCLUSIONS

Our experiments have enabled a better understanding on the roles of applied electric field strength and the frequency of the alternating current (AC). We found that stronger applied fields generate higher field-induced forces on the nanotubes and enhanced the density of the aligned nanotubes. Meanwhile, AC frequency helps to disperse the nanotubes and makes them suitable for better alignment. These approaches have enabled the control of the density and alignment of the CNTs, which was not reported before.

ACKNOWLEDGMENTS

This work is supported by Defense Advanced Research Agency (Contract No: DAAD17-03-C-0115, through Army Research Laboratory), and the U.S. Department of Army (Grant No. W911NF-04-1-0029, through the City College of New York).

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