Dielectrophoretic deposition of carbon nanotubes: The role of field frequency and its dependence on solution conductivity

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ABSTRACT

Dielectrophoretic deposition is ubiquitous in the fabrication of nanoscale devices and, in particular, carbon nanotubes from solutions. The physical properties of the solution play a major role in determining the nanotube patterns deposited using this technique; the non-uniform temperature profile generated by the passage of current can create forces in the solution, which result in agitations and can interfere with the dielectrophoresis force.

The forces that induce movement in the solution, as well as the dielectrophoresis force, all vary with frequency in different manners. In this work, experiments and simulations are used in order to investigate the interplay between the solution conductivity and the frequency of the applied voltage, and their effects on the deposition patterns. We demonstrate that changes in the frequency affect solutions with different physical properties differently: while the frequency directly influences the number of deposited carbon nanotubes when using solutions with low conductivity, it is almost of no consequence in the case of solutions that include surfactants and thus have a higher conductivity.

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1. Introduction

Despite the promising properties of carbon nanotubes (CNTs), reproducibility in fabrication still remains a challenge and a bottleneck for many applications. Dielectrophoresis (DEP) is widely used for the deposition of a CNT or a collection of CNTs between two electrodes [1–4]. DEP offers several advantages: it is performed at low temperatures and enables a high level of control over the location and direction of the deposited nanotubes. Moreover, since solutions containing different types of CNTs are readily available, DEP naturally allows for the fabrication of devices based on specific CNT types.

Nonetheless, DEP results are not always fully reproducible. The role of the various parameters involved such as the amplitude and frequency of the applied voltage, the shape of the electrodes, the duration of experiment, and the properties of the solution, has formed the subject of several previous studies [5–10]. However, important questions still remain.

Current methods for the preparation of aqueous suspensions of CNTs can result in solutions with a wide range of conductivities. Pure CNTs being hydrophobic, the main processes available for suspending them in water involve the addition of surfactant materials [11] or the pre-treatment of the CNTs [12]. Depending on the type and amount of surfactant added, the resulting solution can have different physical properties that affect the DEP process, including electrical conductivity, which can have a significant effect. However, this effect is not fully understood yet and, in fact, has often been neglected.

The application of an alternating voltage between electrodes immersed in a CNT solution leads to a DEP force being exerted on the CNTs. Depending on the permittivity and conductivity of the CNTs and those of the solution, the DEP force will either attract the CNTs to regions where the gradient of the field is the greatest (the gap between the electrodes) or repel them. Another phenomenon occurs due to the heat produced by the current passing through the solution because of the applied voltage. The non-uniform temperature profile leads to a non-uniform distribution of the permittivity and conductivity of the solution, both of which are functions of temperature. Under the applied electric field, a so-called electrothermal force is created, which produces movement in the solution. The magnitude and direction of this force, and the velocity of the resulting movement of the solution, depend on the physical properties of the medium as well as the frequency of the applied electric field [13–15].

A limited number of reports exist on the effect of the movement in the solution during the DEP deposition of CNTs. Using a numerical approach, Lin et al. demonstrated that this movement affects the semiconducting CNTs considerably [16]. In a highly insightful simulation study, Burg et al. adopted a two-dimensional electrokinetic framework in order to evaluate the effect of different forces resulting from the non-uniformity of the temperature profile

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around the electrodes [17]. They demonstrated that the electro-thermal force is the dominant volumetric force on the solutions containing surfactants in the frequency ranges relevant to DEP. Moreover, they showed that the direction of this force depends on the frequency. Although both of the above works investigated the forces affecting the movement of the CNTs at different frequencies, their focus was on the dependence of the movement of the solution on the frequency, rather than its effect on the deposition pattern of the CNTs. Moreover, the simulated frequencies in these works were typically not within the frequency ranges used in DEP experiments on nanotubes.

We previously investigated the simultaneous effects of the electrothermal and DEP forces on CNTs and the resulting deposition pattern at a single frequency for solutions with different levels of conductivity [18]. It was observed that the electrothermal force cannot be neglected in solutions containing surfactants. In addition, the effect of this force does not depend strongly on the percentage of surfactant in stable nanotube solutions; however, it is drastically less pronounced for solutions without surfactant. For solutions with one weight percent (wt%) sodium dodecylbenzene sulfonate (SDBS), which is typical for CNT deposition, the electro-thermal force is the dominant player in the long-range drift of the CNTs, and affects the deposition pattern dramatically compared to the situation in the solution with no surfactant.

An important question here is how the effects of the two main forces – the DEP force and the electrothermal force – evolve with respect to each other as the frequency of the applied voltage is varied. This is a critical issue that requires detailed investigation. In this article, we use experiments and finite element simulations to investigate the interplay between these two forces at different frequencies. We show that there exists a threshold frequency beyond which the number of deposited CNTs bridging the electrodes reduces drastically. For low-conductivity solutions (those made using pre-treated nanotubes), this threshold falls within the frequency range used in practice, whereas in high-conductivity solutions (those including surfactants), it is orders of magnitude higher.

2. Methodology

CNTs are hydrophobic in nature. The two primary methods for suspending nanotubes in water are using surfactants, such as SDBS, or using pre-treatment to functionalize the CNTs. Although both methods result in stable nanotube solutions, the conductivities of the resulting solutions are considerably different.

The concentration of CNTs in a water–surfactant solution depends on the surfactant concentration, which, on the other hand, determines the conductivity of the solution. However, in order to be able to investigate the effect of solution conductivity on the deposition patterns of the CNTs at various frequencies, it is necessary to decouple it from the effect of CNT concentration; one needs to prepare solutions with different conductivities, but identical in other aspects.

To accomplish this, we used a surfactant-free single-walled CNT solution, commercially available from NanoLab Inc. [19], in which the nanotubes are carboxylated prior to suspension in water. The nanotubes had lengths in the range of 1–5 μm and an average diameter of 1.5 nm. The initial concentration of the solution was 1 g/l. The nanotube solution was diluted to the desired concentration. For each sample with a certain concentration of CNTs, two solutions with 0 and 1 wt% SDBS were prepared. Each of the solutions was ultrasonicated for 5 min.

Although the surfactant molecules interact with the CNTs, they do not change the CNT concentration because the CNTs are already suspended. The role of the SDBS here is merely to change the conductivity of the solution. It is noteworthy that, since the SDBS molecules attach to the nanotubes through Van der Waals forces, they do not affect the physical properties of the nanotubes.

The electrodes were fabricated using photolithography and electron beam evaporation, followed by lift-off in acetone, on a p-doped silicon wafer with a 2-μm-thick thermal oxide on the surface. The evaporated metal layers consisted of a 50-nm-thick palladium layer over a 20-nm-thick chromium layer for adhesion. Two types of electrodes were used in the experiments. In the first type, the electrodes were 4 μm in width and had a curved border, with a gap of 4 μm between opposite electrodes; these will be called narrow electrodes from now on. The second type consisted of electrodes 40 μm wide and 4 μm apart from each other; these will be called wide electrodes. The samples were cleaned using acetone, methanol and deionized (DI) water prior to DEP experiments.

The DEP experiments were conducted by applying a 5-V AC signal with a range of frequencies to the electrodes using microprobes. The sample was immersed 3 mm deep into the solution during each experiment. It was then rinsed with DI water followed by blow drying using nitrogen. The samples were imaged using a Hitachi S4700 field-emission scanning electron microscope under 1 kV of primary beam acceleration voltage.

In order to be able to explain the experimental results, 2-dimensional (2D) finite element simulations were performed at
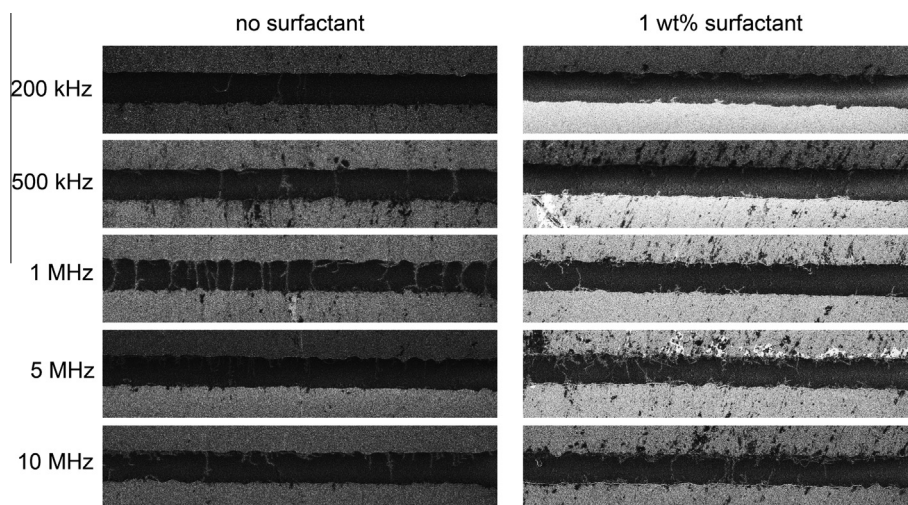

Fig. 1. Scanning electron micrographs of structures fabricated at various frequencies using solutions with no surfactant and 1 wt% surfactant with 2.5 μg/ml of CNTs in water. The gap between the electrodes is 4 μm.