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Controlling SWCNT assembling density by electrokinetics

Haibo Yu [a,d], Sascha Hermann [c], Zaili Dong [a], John Mai [b], Wen J. Li [a,b,*], Stefan E. Schulz [c,d]

[a] Key State Laboratory of Robotics, Shenyang Institute of Automation, CAS, Shenyang, China
[b] Department of Mechanical and Biomedical Engineering, City University of Hong Kong, Hong Kong, China
[c] Center for Microtechnologies (ZFM), Chemnitz University of Technology, Chemnitz, Germany
[d] Department of Back-end of Line, Fraunhofer Institute for Electronic Nano Systems, Chemnitz, Germany

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ABSTRACT

Single-walled carbon nanotubes (SWCNTs) as the colloid in a colloidal solution can be polarized in a non-uniform electric field and experience a net force that is the so-called dielectrophoresis (DEP) force, due to the interaction between the induced dipoles and the electric field. The positive DEP force can be used to position and assemble arrays of SWCNTs. Inversely, the negative DEP force can be utilized to separate SWCNTs in terms of their electronic properties. Moreover, Joule heating generated by the electric field can lead to other electrokinetics forces in the colloidal solution, which give rise to fluidic motion of the solution. Additionally, at low frequencies, the electrical double layer also induces a steady fluidic motion, a phenomenon known as AC electroosmotic flow. These fluidic motion in turn exerts a drag force on the nanotubes. Hence, to controllably assemble SWCNTs using DEP force is a non-trivial task. In this article, the mechanisms of electrokinetics and electrohydrodynamics are systematically analyzed through numerical simulations for a set of parameters that are typically used for assembling SWCNTs between metal electrodes. Finally, experimental results from the frequency-dependent assembly of SWCNTs using this set of parameters are described and discussed. These results show that the density of SWCNTs assembled between electrodes can be varied by controlling the electrokinetics parameters.

1. Introduction

Applications involving single-walled carbon nanotubes (SWCNTs) have been one of the most popular research topics in the fields of nanoelectronic devices, nanoactuator systems, and nanosensors, due to their unique geometric structures, excellent mechanical properties, and extremely high electrical and thermal conductivities [1–5]. The SWCNTs are one-dimensional materials with a diameter ranging from sub-nanometer to several nanometers, and their lengths are usually on the order of micrometers [1]. In terms of their electrical properties, the SWCNTs can be divided into two types, namely metallic and semiconducting [1,2]. The remarkable electronic properties of SWCNTs make them one of the most popular candidates as the building blocks of nanoscale devices [3,4]. However due to their nanoscale characteristics and the diversity in electrical properties in SWCNTs, it is more challenging to fabricate SWCNT-based nanodevices [5–7]. To date several methods have been developed to integrate SWCNTs into nanodevices such as nanomanipulation using an atomic force microscopy (AFM), contact printing and dielectrophoresis (DEP) [8–10].

An AFM is one of the most effective tools for observing the topography of materials and for characterizing the mechanical properties of materials, with a very high resolution on the nanoscale. Imaging analysis is based on the interaction between the AFM tip and the sample. The AFM has also shown its capability to manipulate a SWCNT placed on a substrate [11,12]. With the development of robotic-enhanced operation system based on an AFM, the efficiency of single SWCNT manipulation has increased. The advantage of AFM-based manipulation is that it gives the operator real-time feedback based on images of both the sample and the substrate. In particular, AFM-based nanomanipulation is very high precision. But the typical AFM cannot simultaneously image and manipulate SWCNTs, which limits its application to rapidly fabricate SWCNT-based nanodevices [13].

Over the last decade, most effort has been addressed toward applying DEP to carry out the separation of nanoparticles, nanoassembly and fabrication of nanosensors [14–17]. DEP has shown its great capabilities for aligning SWCNTs in parallel and assembling them between two electrodes in a liquid medium [7,15]. A SWCNT becomes polarized in the presence of an electric field. The polarized SWCNT is driven by a net force that is known as the DEP force, if the electric field is non-uniform. The non-uniform electric field exists when an alternating current (AC) bias is applied between two electrodes. The SWCNT will be driven by the DEP force toward the gradient of the electric field. If the DEP force is positive,

* Corresponding author at: Department of Mechanical and Biomedical Engineering, City University of Hong Kong, Hong Kong. Tel.: +852 3442 9226.
E-mail addresses: wenjli@cityu.edu.hk, wenjungli@gmail.com (W.J. Li).

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the SWCNTs move toward the center between the electrodes and accumulate until they eventually bridge the gap between the electrodes. The DEP method enables the batch manipulation of SWCNTs with a certain degree of uniformity. The large-scale assembly of SWCNTs has been reported using a DEP method [18]. Recent efforts to use DEP on SWCNTs have primarily focused on the large-scale assembly of SWCNT nanodevices by capacitive coupling between the silicon substrate and the microelectrodes [18,19].

However, the application of DEP in manipulating SWCNTs has been hampered by several main challenges. One of them is that the SWCNTs are essentially insoluble in most common solvents [20]. The usage of surfactants to disperse nanotubes results in changes in their surface conductivity [21]. Moreover, the SWCNT materials are usually heterogeneous, which lead to differences in polarization of SWCNTs. Furthermore, the electric field gives rise to an electrothermal flow and an electroosmosis flow, which will exert a viscous drag forces on nanotubes. These drag forces are also frequency dependent and can become dominant below a certain applied frequency.

In this work, the DEP force acting on metallic and semiconducting SWCNTs (m-SWCNTs and s-SWCNTs) is first analyzed. The viscous forces induced by the electrothermal flow and the electroosmosis flow is formulated. Comparisons of the DEP force with the electrothermal force and the electroosmosis force are discussed with respect to variations in the DEP frequencies, the applied voltages, and the nanotube lengths. Finally, the importance of frequency-dependent assembly is studied experimentally.

2. Electrokinetics and electrohydrodynamics

2.1. Dielectrophoresis

A polarizable particle in an electric field forms an induced dipole inner the particle [22]. The DEP force on the particle arises from the interaction of the induced dipole and the non-uniform electric field. The polarization of SWCNTs dominates the electrophoretic and DEP forces. The positive DEP force moves the particle toward a region with a high gradient of electric field strength. Conversely, the negative DEP force moves the particle toward regions with a low gradient of electric field strength. The direction of the DEP force, and thus the direction of the particle movement, depends on the difference between the permittivity of the particle and solution medium. The frequency-dependent polarization factor is related to the conductivity and permittivity of the SWCNT as well as the conductivity and permittivity of the suspending medium.

For a rod-shaped, ellipsoidal particle with its long axis parallel to the electric field lines, the time-averaged DEP force is expressed as [23–26]

$$\langle F_{\mathrm{DEP}} \rangle = \frac{\pi abc}{3} \varepsilon_1 \operatorname{Re}\{f_{\mathrm{CM}}\} \nabla \left| E \right|^2 \tag{1}$$

where a, b, and c are the half lengths of the major ellipsoidal axes, respectively. The direction and magnitude of the DEP force are attributed to the polarizability ratio of particle, which can be described by the Clausius–Mossotti (CM) factor [24]

$$f_{\mathrm{CM}} = \frac{\varepsilon'_p - \varepsilon'_m}{\varepsilon'_m + (\varepsilon'_p - \varepsilon'_m) L_{||}} \tag{2}$$

where ε'_m and ε'_p are the complex permittivity of the suspending medium and particle, respectively. The complex permittivity of the particle can be written as

$$\varepsilon' = \varepsilon - i\frac{\sigma}{\omega} \tag{3}$$

where ε is the permittivity, σ is the conductivity, and $\omega = 2\pi f$ is the angular frequency of the AC electric field. In any given solution, the magnitude of the DEP force on a constant-sized particle mainly depends on the complex permittivity.

$L_{||}$ is the depolarized factor related to the length and diameter of the SWCNT, and E is the electric field. For a SWCNT, the length-to-diameter ratio is very large. Therefore, the SWCNT can be modeled as a long, thin rode, which shows highly polarization along the long-axis direction. The depolarization factor can be written as [25]

$$L_{||} \approx \frac{d^2}{2l^2 e^3} \left[\ln\left(\frac{1+e}{1-e} \right) \right] \tag{4}$$

where d is the diameter and l is the length of the SWCNT, respectively, and

$$e = \sqrt{\frac{(1-d^2)}{l^2}} \tag{5}$$

In the low frequency region, the real part of the CM factor relies mainly on the conductivities of the particle and the suspending medium. Conversely, in the high-frequency region, the real part of the CM factor is dominated by the permittivities of the particle and the suspending medium.

2.2. Electrothermal flow

The hydrodynamic behavior of the suspending medium must be taken into account. Due to the conductivity of the solution, the electric field gives rise to Joule heating [27]. Local heating of the fluid causes gradients in the conductivity and permittivity. As a result, a body force is generated on the fluid, which induces an electrothermal flow. The flow of the fluid imposes an electrothermal force on the SWCNTs. The time-averaged electrothermal force can be written as [22]

$$\langle f_e \rangle = \frac{1}{2} \operatorname{Re}\left[\frac{\sigma_m \varepsilon_m (\alpha - \beta)}{\sigma_m + i\omega\varepsilon_m} (\nabla T \cdot \mathbf{E}_0) \mathbf{E}_0^* - \frac{1}{2} \varepsilon_m \alpha \left| \mathbf{E}_0 \right|^2 \nabla T \right] \tag{6}$$

with the permittivity term $\alpha = \partial\varepsilon_m / \partial T / \varepsilon_m \approx -0.004\,\mathrm{K}^{-1}$ and the conductivity term $\beta = \partial\sigma_m / \partial T / \sigma_m \approx 0.02\,\mathrm{K}^{-1}$ [28].

For a low Reynolds number system, the velocity field of the fluid can be obtained by solving the incompressible Navier–Stokes equation. As an approximation, the fluid velocity can be calculated by comparing the electrothermal force with the viscous force, which can be expressed as $v_e \approx \left| f_e \right| l_C^2 / \eta$ [29]. l_C is the characteristic length of system.

2.3. AC electroosmosis flow

In the lower frequency ranges, AC electroosmosis flow arises, which is due to the tangential electric field on the electrical double layer. The migration of charges generates a drag flow in the fluid. As a result, the electroosmosis flow results in a slip velocity at the electrolyte–electrode interface. The fluid velocity depends both on the applied potential and the frequency. If the characteristic thickness of the double layer is far smaller than the vertical characteristic length, the time-averaged AC electroosmosis slip velocity can be written as [22]

$$v_{aceo} = \Lambda \frac{\varepsilon_m V^2}{8\eta x} \frac{\Omega^2}{(1+\Omega^2)^2} \tag{7}$$

The non-dimensional frequency is given by [22]

$$\Omega = \Lambda \frac{\omega \varepsilon_m \pi x}{2\sigma_m \lambda_D} \tag{8}$$

where x is the distance from the center of the electrode gap, the factor Λ is defined as a ratio between the capacitance of the Stern layer and the overall double layer capacitance, which depends mainly