The study of particle-particle interaction and assembly under the influence of dielectrophoretic force experienced between carbon microelectrodes

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Abstract

Electrokinetic microassembly is used to position microparts via dielectrophoresis and electro-osmosis. Present work studies experimental conditions for creating latex particle agglomerates such as “pearly chains”. The work discusses one of the approached to lock particles in the position after the electric field is removed. Particle-to-particle interactions such as electro-rotation and formation of particle chains parallel and perpendicular to the field lines are explored.

Keywords: Microassembly, Dielectrophoresis, Particle-particle Interactions, Electro-rotation.

1. Introduction

Manufacturing techniques can be broadly separated into reductive and additive technologies. Among the latter fabrication methods one of the promising directions is the that of dielectrophoretic electropolymerization (DEP EP) [1], where microparticulates ranging from organic and inorganic particles to biological cells can be guided towards the microelectrodes by dielectrophoretic force and then be permanently captured and attached to the surface by the thin layer of conducting electrodes such as polypyrrole (PPy) that is electrodeposited from the solution once the particles are already attracted to the microelectrodes.

Dielectrophoretic (DEP) force is exerted on non-conductive particles placed in non-uniform electric field [2]. This force F_{DEP} on a spherical particle can be calculated by first finding the dipole moment of the electrostatically polarized sphere in the uniform field and then analysing the force on that dipole in the actual non-uniform field. This approach yields the value of the DEP force:

$$F_{DEP}(\omega) = 2\pi r^3 \varepsilon_m \mathrm{Re}[K] \nabla E^2_{rms} \quad (1)$$

where r is the radius of the particle, ε_m is electrical permittivity of the media, ω is the frequency of the applied AC signal, Erms is the root-mean-square of the electric field, Re [K] is the real part of factor K. The Claussius- Mossotti factor K is given by:

$$K = (\varepsilon^*_p - \varepsilon^*_m)/(\varepsilon^*_p + 2\varepsilon^*_m) \quad (2)$$

where complex permittivity of particle (subscript p) and medium (subscript m) are calculated as given in Eq. (3):

$$\varepsilon^* = \varepsilon - j\,\sigma/\omega \quad (3)$$

where σ is electrical conductivity (of either particle or medium, depending on the subscript) and j is the unit imaginary number.

If the Claussius-Mossotti factor K is positive, then the particle will experience positive DEP force (pDEP) and will be attracted to the microelectrodes, while negative DEP (nDEP) will result in particles repelled from the electrodes.

The described physical picture works well for low particle concentration when the main interaction is between the individual particles and the imposed electric field. However, as particle concentration grows, particle-to-particle interaction start to play increasing role. For example, when several particles, that each have charge separation due to the induced polarization, come to the close vicinity to each other, they will reorient themselves to have charges of the opposite signs on nearby particles attract each other and thus chains of connected particles, so-called “pearl chains”, are produced. Such chains, formed with dielectric latex beads or biological cells [3, 4] have transitory nature and are broken apart when the signal is turned off, since van der Walls forces are not strong enough to prevent electrostatic instability.

Formation of long particle chains spanning the gap between the electrodes can be a useful micromanufacturing technique if only these chains can be somehow fixed in place after their formation. For example, when the chains are fixed in place, at least temporarily, electroactive conducting polymers, such as PPy can be deposited over the particles [5], forming a conducting bridge between the interdigitated electrodes (see Fig. 1a), forming a basis for a sensor, a transistor, or other electronic devices.

While some qualitative analysis was made for small-size pearl chains (two to four sequential beads), [6], only a general phenomenological qualitative prediction is available for chaining force and stability of longer chains [7].

The present work is focused on study of experimental conditions for production of pearl chains that can be formed across the gap between the interdigitated microelectrodes and on the methodology for temporary fixing these chains in place (subsequent electropolymerization of the deposited chains is outside the scope of the present study). Majority of the previous studies of pearl chain formation was

performed with metals electrodes (gold, platinum or similar) [8], and these studies were limited by hydrolysis that takes place at low frequencies. We are utilizing C-MEMs process [9] to construct carbon microelectrodes that have wider stability window than the metal electrodes and will not experience hydrolysis over wider range of applied frequencies and voltages. We are also reporting on experimental conditions for production of field-perpendicular chains (i.e. parallel to the long axis of electrodes – Fig.1b) that hitherto were thought to apply only to dissimilar particles [10]. Finally, we discuss the electrorotation (Fig.1c) [11] of the particle agglomerates. The electro-rotation process might be utilized for mixing and homogenization of the fluid sample or reagents. The summary of studied phenomena, including two types of pearl chaining and electrorotation is presented in Fig. 1.

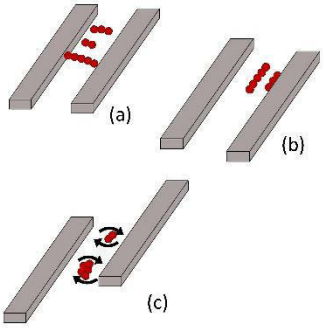

Fig 1. Particle-particle interactions studied in the present work include (a) pearly chains along the field lines, (b) pearly chains perpendicular to the field lines, and (c) electrorotation of particle aggregates.

2. Materials and Methods

2.1. Fabrication

The carbon electrode design used in this work consists of a planar interdigitated array (Fig. 2). The electrode array fingers have the width of 15 μm and have electrode-to-electrode spaces of 25 μm. The carbon lines connecting the planar interdigitated array to the conductivity pads have a width of 770 μm. A mask printed on acrylic (CAD/Art Services, Inc., OR, USA) contains patterns for 20 individual chips. A 100 mm diameter single-crystal Si wafer was used as the substrate. Spincoater P6700 (Specialty Coating Systems, INC., IN, USA) used two step sequence of spin at 500 rpm for 10 seconds followed by a spin at 2750 rpm for 30 seconds, to deposit 25nm layer of SU-8 2025 negative photoresist (MicroChem, MA, USA). The resist underwent soft bake on a Programmable Hotplate HP61 (Torrey Pines Scientific, CA, USA) by ramping from room temperature to 95 ºC at a rate of 120 ºC/hr, then maintaining at 95 ºC for 5 minutes, and lastly allowing natural cooled back to room temperature. Afterwards, the flood exposure of UV radiation with a dose of 2 MW /cm² for 60 seconds was used to transfer mask pattern to the resist. Following the UV exposure, the wafer substrate was soft baked again with the same temperature regime as that prior to the exposure. Afterwards, the substrate was placed in a SU-8 developer for 5 minutes in order to develop the photoresist. Taking the wafer out of the developer bath, it was cleaned with acetone and isopropanol to remove the excess photoresist. Optical inspection was performed to verify the fidelity of the transferred pattern. After the SU-8 pattern of the electrodes was produced on the wafer, it was placed in the quartz tube of the furnace to perform pyrolysis in order to convert SU-8 resin into the carbon. The furnace was purged with nitrogen gas at room temperature for 1 hour at 0.5 SCFH. The temperature regime consisted of the ramp from room temperature to 300°C for 5 hours, then holding at 300°C for 2 hours, ramp to 900°C during 3 hours, holding at 900°C, for an hour, and allow natural cooling to room temperature overnight. Each wafer contains 20 arrays of interdigitated electrodes. Diamond scribe is used to dice the wafer into individual chips.

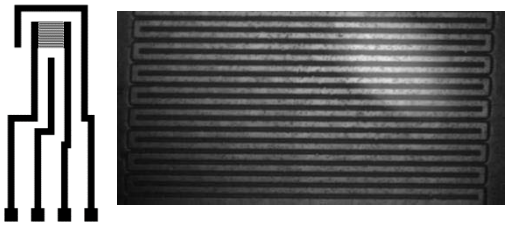

Fig 2. (a) Sketch of the Interdigitated Electrode Array (electrode fingers width is 15 microns) (b) Close up image of the electrode array with polystyrene microbeads.

2.2. Experimental Setup

Carboxylated polystyrene microbeads 2 μm in diameter (Magsphere Inc., Pasadena, CA) were suspended in deionized (DI) water. The conductivity probe (Hanna Instruments) was employed to measure the conductivity of the DI water which was found to be σ_m = 6.00 x10^{-6} S/m. A small droplet of the microbead suspension was placed with a pipet onto the interdigitated electrodes. An Eclipse LV-UDM Universal Design Microscope (Nikon Instruments Inc., Melville, NY) was utilized to observe the electrokinetic phenomena. An indium solder was used to attach wires to the carbon contact pads of the electrode chips. The wires were connected to the Function Generator DS345 (Stanford Research Systems, Sunnyvale, CA) that applied AC voltage to the chip. The dynamics of the microbead movements can be observed on the computer screen and stored as a series of images taken by a SPOT RT KE CCD camera (Diagnostic Instruments Inc. Sterling Heights,MI) attached to the microscope. The experimental setup is presented in Fig. 3.

3. Computational Modeling

Fig. 3. Experimental set-up.

The software COMSOL Multiphysics 5.3 © (COMSOL Inc., Burlington, MA) was used to simulate the electric fields produced in the solution when

varying frequencies are applied onto a carbon electrode array.

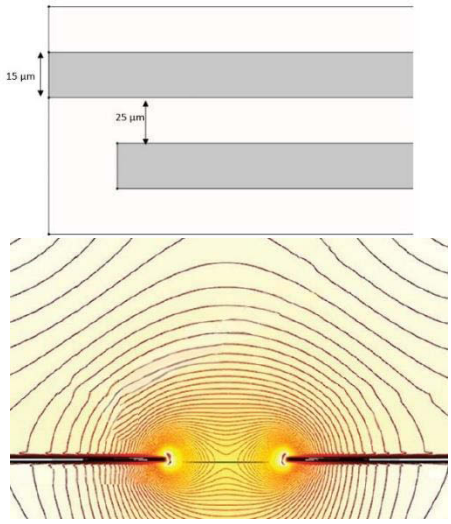

Fig. 4. Pair of the electrodes – top view (top) and the simulation results for the electrical field distribution with equipotential lines – the side view (bottom).

The electrode geometry drafted for the simulation reflects the actual interdigitated electrode array (IDEA) from the carbon electrodes used for experimentation. The IDEA contains 50 fingers that are 15 microns in width with inter-electrode gaps of 25 microns (see Fig. 4 top).

The computational model displays the electric field lines between the electrodes (Fig.4 bottom). It can be seen that the largest gradient of the field exist the electrode edges and thus dielectrophoretic forces will be highest in these regions. Indeed, experimental observations confirm that result.

Relative permittivity of polystyrene was takes as ε_p=2.5 and its electrical conductivity as σ_p =1.0x10^{-3} S/m [12, 13]. Relative permittivity of water was taken as ε_m=78. These parameters are used to calculate frequency-dependents Claussius- Mossotti factor K (given by the Eq. 2 above). The calculated results are presented on the plot in Fig. 5. The predicted cross-over frequency (at which the beads are neither attracted nor repelled from the electrodes) is around 2 MHz.

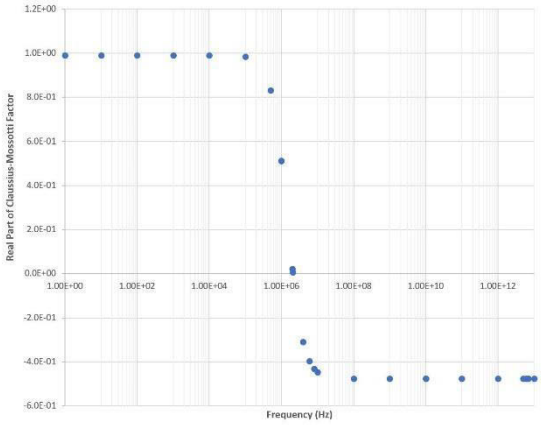

Fig. 5. Plot of the real part of the Claussius-Mossotti Factor predicting the crossover frequency.

4. Results and Discussion

The observed crossover frequency of 1.25 MHz is somewhat lower than the estimated 2MHz. The deviation from the predicted crossover frequency is likely due to the difference between the assumed (not measured experimentally) surface bead conductivity and the actual latex bead conductivity.

At the applied frequency of 300kHz and peak to peak voltage V_{pp}=1V individual beads were seen to rotate. As frequency was increased to 1MHz and the bias was increased to Vpp = 6V besides electro-rotation, particles were seen to coalesce into larger clusters and continue their rotation as seen in Fig. 6. It is important to note that prior theories of electro-rotation imply that electro-osmosis makes a significant contribution to electro-rotation [11]. However, present experimental results with deionized water and negligible ionic concentration demonstrates that the dominant component of electrorotation is charge re-distribution on the beads and the created dipole moments that provide necessary torque to enact the electrorotation of first, individual beads, and gradually – increasingly larger bead aggregates.

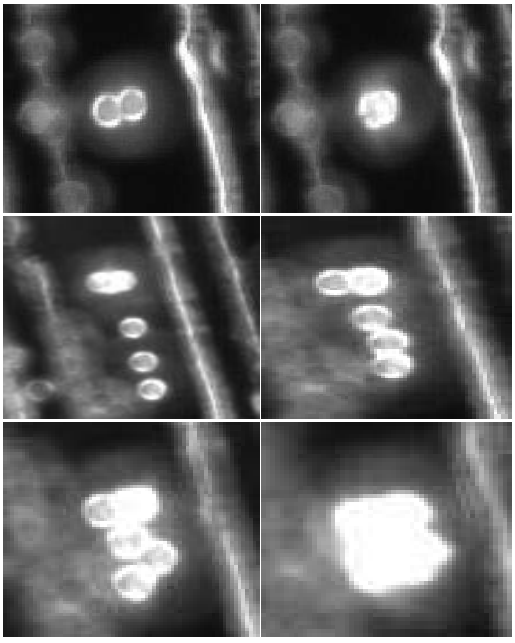

Fig.6. A sequence of dark field micrographs of electro-rotating and aggregating particles (2 micron diameter) at the applied frequency of 1 MHz and Vpp = 6V.

Around 300 kHz (V_{pp}=6V) particle-to-particle interaction also lead to the formation of pearly chains along the field lines (Fig.7 left). If the field is turned off and the chip is left to dry, the chains will stay in place (Fig. 7 right). When the fresh solution is added, the chains stay in place due to adhesion enacted by the surface tension force applied during the drying process. This “fixing-by-drying” of the pearly bridges can be potentially used to add electroactive monomer solution to electrodeposit conducting polymers such as polypyrrole to permanently attach and cover beads with the thin conductive polymer layer, as was demonstrated previously [1].

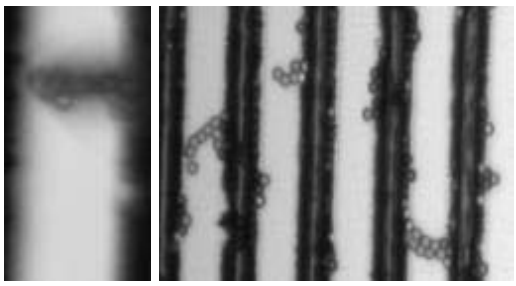

Fig. 7. (left) Beads lined into the pearly chain along the field lines at 300 kHz (V_{pp}=6V); (right) After the chip is dried and the signal is removed, the beads adhere to the chip, forming fixed patterns of beads.

At the frequency of 1.2 MHz the positive DEP directed beads to the electrodes, while particle-to-particle interaction promoted the chain formation. Interesting phenomenon of chain formation perpendicular to the filed lines (i.e. parallel to the electrodes) was observed as pearly particle chains formed at the electrodes continued to grow by attracting neighboring beads to roll along the electrode and attach themselves to the chain. Fig. 8 presents the latex beads chain at the electrode growing as captured in the four sequential frames. The growing chain is

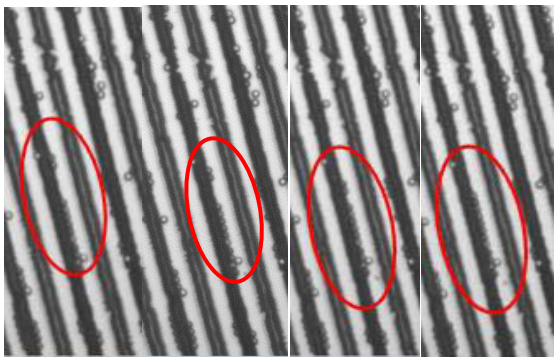

Fig. 8. Bead chains growing at the electrode (1.2MHz, V_{pp}=1V).

circled in red. Prior to this study, the literature reported on formation of perpendicular-to-field chains only for dissimilar particles [8].

5. Conclusions

The present study performed experimental investigation of the particle-to-particle interaction of the latex beads suspended in the deionized water over the interdigitated microelectrodes. The AC fields was varied from several Hz to 10 MHz and the applied peak-to-peak voltage varied from 1 V to 6 V. Experimental conditions for formation of so-called “pearly chains” aligned along the field lines, perpendicular to the field lines, and for electro-rotation of the beads and bead agglomerates was reported. “Fixing-by-drying” technique for locking bead patterns in place after the electric field is removed was discussed as well.

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References

[1] V. H. Perez-Gonzalez et al., “A novel micro/nano fabrication process based on the combined use of dielectrophoresis, electroosmotic flow, and electrodeposition for surface patterning,” Journal of Micromechanics and Microengineering, 2015; 25(11): 115007.
[2] H. Morgan, H., Green, N. G., AC electrokinetics: colloids and nanoparticles, Research Studies Press: 2003.
[3] S. Fraden et al., “Electric-field-induced association of colloidal particles,” Physical review letters, 1989; 63(21): 2373.
[4] J. L. Griffin et al., ”Pearl chain formation across radio frequency fields,” Nature, 1970: 226(5241): 152.
[5] V. H. Perez-Gonzalez et al., “Dielectrophoresis-assisted electroconductive polymer-based fabrication of high surface area electrodes,” Electroactive Polymer Actuators and Devices (EAPAD): 2014, 905628.
[6] H. Moncada- Hernandez et al., “Theoretical and experimental examination of particle–particle interaction effects on induced dipole moments and dielectrophoretic responses of multiple particle chains,” Electrophoresis, 2014; 35(12-13): 1803-1813.
[7] T. B. Jones et al., Electromechanics of particles, Cambridge University Press: 2005.
[8] O. D. Velev et al., “On-chip micromanipulation and assembly of colloidal particles by electric fields,” Soft Matter, 2006; 2(9): 738-750.
[9] V. Ho et al., “Novel fabrication technology for three-dimensional high surface area pyrolized structures,” Sensors and Smart Structures Technologies for Civil, Mechanical, and Aerospace Systems: 2010, 76472F.
[10] W. M. Arnold, “Particle patterning using fluidics and electric fields,” IEEE Transactions on Dielectrics and Electrical Insulation, 2008; 15(1).
[11] C. Grosse et al., “Theory of the low-frequency electrorotation of polystyrene particles in electrolyte solution,” The Journal of Physical Chemistry, 1996; 100(5): 1771-1778.
[12] N. G. Green et al., “Dielectrophoresis of submicrometer latex spheres,” J. Phys. Chem. B, 1999; 103, 41–50.
[13] M. P. Hughes, “Dielectrophoretic behavior of latex nanospheres: low-frequency dispersion,” J. Colloid. Interf. Sci., 2002; 250: 291–4.