DEP MICROACTUATION OF LIQUIDS

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ABSTRACT

We report rapid actuation of nanoliter to microliter water volumes using the dielectrophoretic (DEP) force. This micro-electromechanical mechanism for manipulating, transporting, and metering liquids harnesses strong, non-uniform rf electric fields created by co-planar electrodes with 30 to 100 µm feature size patterned on insulating substrates. Electrolysis is avoided by spin-coating the electrodes with ~10 µm of polyimide. Transient liquid velocities exceed 25 cm/s, and droplets down to a few nanoliters in volume are formed in <30 ms. Because water responds rapidly, controlled DEP actuation can be achieved by very short bursts of rf voltage. An important benefit of minimized exposure of conductive liquids to strong rf fields is reduced Joule heating. A new microfluidic scheme for the "laboratory on a chip" is proposed, based on high-speed, programmable manipulation of discrete nanodroplets on smooth substrates.

INTRODUCTION AND BACKGROUND

To realize the "laboratory on a chip", microfluidic systems are needed to control and manipulate small liquid inventories. Schemes for this purpose include micro-channels that exploit microcapillarity **[1,2]**, electrophoretic and electro-osmotic pumping **[3,4]**, and voltage- or light-mediated surface wetting **[5,6]**. In contrast to these, liquid DEP exploits the ponderomotive force to create hydrostatic equilibria, where dielectric liquids are configured by non-uniform electric fields **[7,8]**. On the microscale, liquid DEP offers the unique capability of very rapid voltage-controlled manipulation of water volumes ranging from ~10 microliters down to ~1 nanoliter (or less) using easily fabricated, co-planar electrode structures.

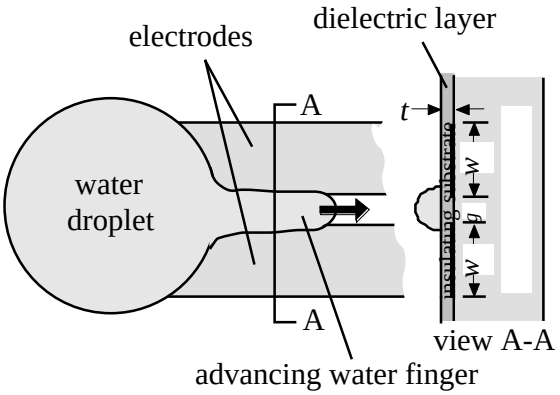

Figure 1. *Basic co-planar electrode structure (top and cross-sectional views) with finger projecting from initial droplet. Inset A-A shows the semi-circular cross-sectional profile of water finger at high frequency. Not shown to scale*

The dielectrophoretic force attracts dielectrics including liquids to regions of strong electric field. Fig. 1 depicts field-induced, transient flow of water in a simple electrode structure consisting of parallel, co-planar strips. For effective actuation, the frequency of the voltage must exceed a minimum value which is determined by the conductivity of the water and the capacitance per unit area of the dielectric coating covering the electrodes. For DI water of conductivity ~10^{-4} S/m, this frequency is ~60 kHz, a value successfully predicted using a simple RC circuit model **[9]**. When voltage is applied at sufficient frequency, a finger of water having the compact, semi-circular cross-section shown in view A-A, extends from the microliter-sized droplet along the axis of the electrodes. Fig. 2 contains selected video images from a test where the voltage was turned up by hand. If the rf field is rapidly applied, the behavior is similar, but the finger moves considerably faster. When the field is removed, the capillary-jet instability rapidly breaks up the liquid into droplets distributed along the length of the flow structure.

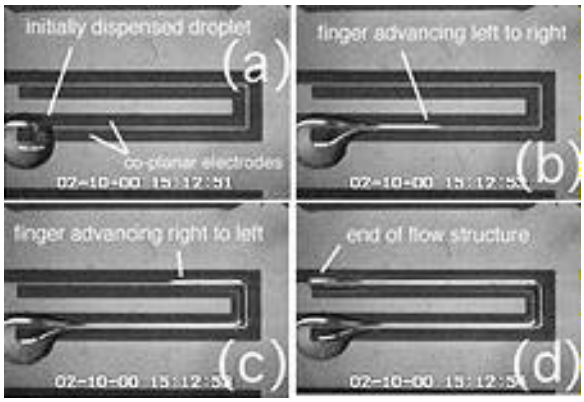

Figure 2. *Selected video images of a "wall-less" DEP flow structure: w = 1000 µm, g = 100 µm, t = 10 µm; DI water (~10^{-4} S/m); voltage at ~100 kHz is turned up by hand to ~700 V-rms. (a) Voltage off. (b) Voltage on: finger moving to right. (c)) Voltage on: finger has rounded two 90º turns. (d) Voltage on: finger has reached end of structure and transient flow is strating to slow down.*

NANOLITER DROPLET GENERATION

The basic parallel strip geometry of the flow structure demonstrated in Fig. 2 can be converted into a droplet generator

by making the electrode strips much narrower, and then terminating them in a pair of closely spaced half-circles. The sequence of video images in Fig. 3 shows a DEP droplet generator in operation. Upon application of rf voltage, a finger of water emerges from the large (~10 µliter) droplet and moves rapidly from right to left. On reaching the end, the finger inundates the circle with water and quickly accumulates an essentially hemispherical mass having volume ~60 nanoliters. As long as the rf field is turned on, this droplet remains attached to the liquid in the flow structure. The DEP force effectively constrains liquid all along the contact line with the polyimide layer and suppresses the capillary instability **[10]**; when the field is removed, this instability pinches off the liquid and isolates the droplet.

Multiple droplets may be generated at high speed if the bisected circles are arrayed at intervals along a flow structure. The sequence of video frames shown in Fig. 4 shows operation of such a structure with four in-line droplet forming circles. Rf voltage (at 700 kHz) is turned up rapidly by hand to ~700 V and then quickly removed in a lapsed time less than ~1 s. In response, a droplet accumulates atop each of the circles. Each of these droplets has a volume of ~4 nanoliters.

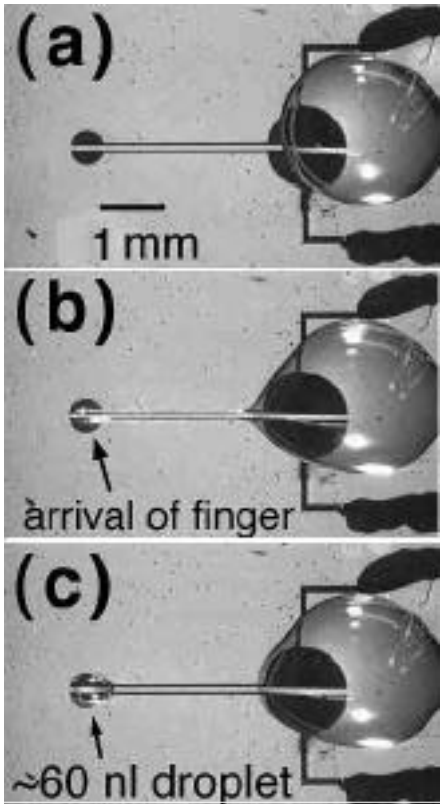

Figure 3. *Co-planar strips terminated in semi-circular electrodes for droplet formation. (a) Voltage off; initial ~10 µliter droplet at right. (b) Voltage on; finger has reached semi-circular electrodes at left. (c) Voltage off; isolated ~60 nanoliter droplet at left; flow structure is completely drained.*

When the voltage is applied using a dc-actuated electromechanical relay, a rise-time on the order of ~5 milliseconds is achieved for the rf electric field. At such speeds, the transient dynamics of finger motion and droplet formation are far too fast to be captured using a conventional video camera operating at 30 frames per second (fps). We now use a high-speed motion analysis camera (Kodak Ektapro HS™) operating at 1125 fps. Analysis of the videos reveals that the velocity of the leading edge of a finger moves as fast as ~25 cm/s. In the droplet generator structures, the leading edge

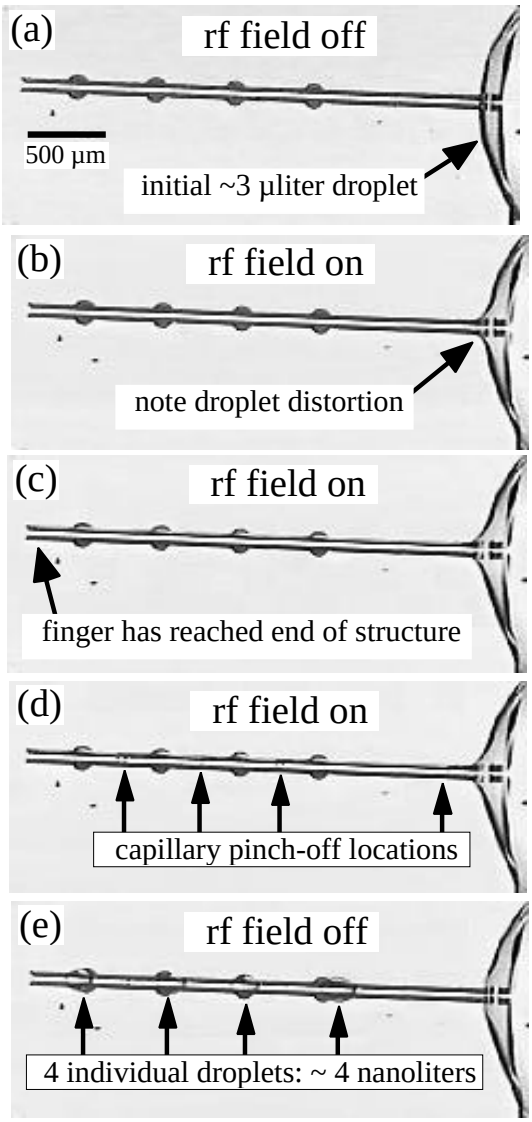

Figure 4. *Multiple droplet generation structure with initially deposited ~3 µl droplet at right. (a) Rf field off. (b) Field on; droplet is distorted. (c) Field on; finger has reached end of structure at left. (d) Field going off; liquid is pinching off between each of the circles. (e) Field off; 4 hemispherical droplets ~4 nanoliters in volume are formed.*

of the finger first passes a droplet-forming circle, and then the circle fills in a time ranging from ~4 to ~12 ms, depending on its distance from the initial (large) droplet. With the relay, the fall time of the rf field is ~3 ms, and the capillary instability separates the droplets in about this same amount of time. Thus, at the present time, we do not know whether or not the capillary instability might actually occur more rapidly.

It has been found that droplet formation is not well-controlled for narrower flow structures. Fig. 4e shows this problem for a flow structure with a total width of ~130 μm, i.e., w = 50 μm and g = 30 μm. In this frame, the four droplets are neither accurately centered upon the circles nor uniform in volume. Several related factors may contribute to this behavior. First, if the circles are too far apart, the capillary jet instability may initiate breakup of liquid in the flow structures at multiple locations between droplets. Second, there will likely exist a momentum imbalance as the masses of water arriving from opposite directions are drawn in by capillarity. To improve droplet uniformity, the locations of capillary pinch-off must be better controlled. Patterning the circles closer together and interposing narrow, hydrophobic strips midway between the circles to promote pinch-off at preferred locations along the flow structures might be effective.

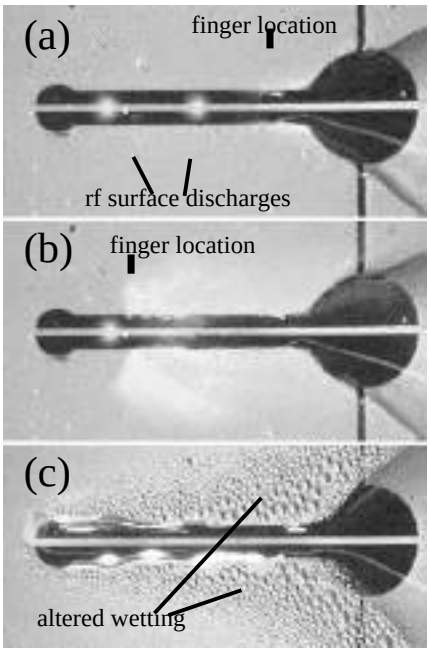

Figure 5. *Rf surface discharge in flow structure. (a) Finger advancing from right to left and two rf discharge sites. (b) Quenching of first discharge by arriving finger; note steam generation. (c) Voltage off; permanent alteration of wetting properties of polyimide is evident.*

A further objective, of great importance for any practical application of microDEP in the laboratory on a chip, is to scale up the number of droplets produced at a time, while simultaneously scaling down the volume per droplet. Though reasonably confident that at least ten sub-nanoliter droplets can be formed at a time with a structure similar to that shown in Fig. 4, we do not know either the minimum droplet volume or the maximum number that can be formed by DEP.

DISCUSSION

The transient fluid dynamics of small liquid masses resting on a substrate and driven by strong, non-uniform electric fields have not been studied extensively, if at all. In particular, the strong interactions we observe that involve the DEP force, capillarity, and surface wetting are not well-understood. For example, there is some evidence suggesting that the liquid/solid wetting angle must lie within a limited range of values for successful DEP actuation. The wetting angle of water on polyimide is ~75º, and this combination shows good DEP microactuation; on the other hand, coating the polyimide with either amorphous Teflon®, with contact angle 105º, or agarose, a good wetting agent, gives poor results. Furthermore, surface rf discharges have a strong influence on wetting conditions. Refer to Fig. 5, which shows visible surface discharges between the electrodes, and subsequent rather dramatic changes in the wetting. This particular structure failed to work when it was tested a second time. While visible discharges like those captured in these video images are quite rare, observations with an image intensifier camera suggest that lower intensity rf discharges actually may be rather common. Some electrode structures, particularly droplet generators, work several times before failure (the record is 9 times), while others work only once or not at all. The specific causes for our variable "yield" rates and for the unpredictable failure of fabricated structures are not known, but there seems to be a correlation with surface wetting.

A very serious, practical problem is that, for all the experiments reported to date, the voltages required to achieve DEP actuation have been too high: typically ~500 to ~700 V-rms for structures with electrode spacing g ~ 100 μm and dielectric thickness t ~ 10 μm of spin-coated polyimide. Such voltages are likely to become particularly troublesome at operating frequencies above 10^2 kHz. The most effective strategy for reduction of the required voltage is to use thinner dielectric coatings on the electrodes. Thinner polyimide coatings have been tried but with unsatisfactory results, possibly due to deficiencies in the spin-coating procedures used. Many other candidate coatings are being considered, though the search for an ideal material is challenging because of the dual requirements of retaining good dielectric breakdown strength and controlling surface wetting properties.

A second way to lower the voltage requirement is to reduce the feature size of the electrode structures, particularly the inter-electrode spacing g. We anticipate no problems in reducing the spacing from the present ~30 microns to ~10 microns, or perhaps smaller. Another benefit of smaller structure size will be the capability to reduce droplet size below 1 nanoliter. While the ultimately achievable lower limit on droplet formation size is not yet established, we are confident that water droplets below 1 nanoliter can be formed using DEP.

An important benefit to be anticipated if the required voltage can be reduced to ~100 V is that rf surface discharges and the attendant surface damage problems probably should be ameliorated.

CONCLUSION

For droplet volumes ranging from microliters down to nanoliters and planar microelectrodes with gaps from ~10 to ~100 μm, dielectrophoresis provides a unique, controllable force for the manipulation of small water volumes. Using DEP, we have demonstrated field-mediated formation of multiple droplets of volume ~4 nanoliters in less than ~30 milliseconds. One may envision a smart microfluidic technology based on liquid DEP microactuation with very interesting potential applications in the "laboratory on a chip". Such a scheme might be called a "nanodroplet switchyard", where microliter sample inventories, dispensed manually or otherwise on a substrate, are manipulated, metered, and divided down to nanoliter droplets for subsequent transport, diagnostics, mixing, separation, and/or other processing operations. Droplets formed by DEP could probably be transported on a chip using the scheme proposed by Washizu **[11]**. The co-planar electrodes might be individually addressable, making it possible to direct liquid masses along different paths or channels on the substrate. Because of the rapid response of the liquid, the rf electric field could be applied in short bursts to the co-planar electrodes. The operation of such a system would be fully programmable and automated.

The "nanodroplet switchyard" scheme represents a unique version of the "laboratory on a chip". First of all, the open, planar structures are easy to fabricate and clean. They might also be cheap enough to be disposable. There will be no problems associated with leakage, priming, or clogging. Using the strategy of dividing the samples into discrete droplets, there should be little likelihood of inter-sample mixing or cross-contamination, an important consideration in the case of critical processes and biological assay protocols. A further potential application for liquid microDEP might be to provide a simple means to "interface" other closed-channel, microfluidic devices with macroscopic systems **[12]**.

DEP liquid actuation on the microscale involves a complex interplay of phenomena: dielectrophoresis, capillarity and wetting, transient fluid dynamics, joule heating, and rf discharge. Many questions are unanswered. We know neither the lower limit on droplet formation size nor the upper limit on electrical conductivity for effective DEP actuation of aqueous media. Major challenges to development of the droplet switchyard include amelioration of Joule heating, scaling down electrode structure sizes to facilitate sub-nanoliter droplet formation, reduction of the required voltage, and improvement to the dielectric coatings to make them less susceptible to the rf surface discharges which can rapidly alter surface wetting.

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