DROPLET MANIPULATIONS BY DIELECTROWETTING: CREATING, TRANSPORTING, SPLITTING, AND MERGING

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ABSTRACT

This paper presents four droplet operations (creating, transporting, splitting and merging of droplets) actuated by dielectrowetting (not electrowetting), which fundamentally constitute a backbone of droplet-based digital microfluidics. These operations are successfully achieved for both non-conductive (dielectric) and conductive (DI water with/without surfactant) fluids in an open environment on a single plate. All these features are attributed to the dielectrowetting nature of superspreading. By contrast, electrowetting works only with conductive fluids, commonly in the two-plate (hundreds microns gap) channel environment. Particularly, splitting and creating by electrowetting have never been realized on a single plate. As for the present device, these successful operations significantly enhance digital microfluidics to work for a wide range of fluids, to simplify the required physical structures, and to increase the handing fluid volumes (otherwise limited by the two-plate gap).

INTRODUCTION

Four droplet operation units (creating, transporting, splitting and merging) are fundamental to digital microfluidics, which correspond to dispensing, pumping, volume controlling and mixing in the microchannel counterpart. These operations were well demonstrated previously by electorwetting on dielectric (EWOD) digital circuit [1]. Such device generally consists of two parallel plates with several hundred micrometers spacing. The droplets are sandwiched in the parallel-plate channel, where the top and bottom walls are coated by dielectric and hydrophobic materials in sequence, such as parylene and Teflon, respectively. The wetting property of conductive liquid can be changed by energizing the electrodes on the bottom plate. Since the contact angle of droplet is locally decreased, the asymmetric meniscuses induce a pressure difference inside. As a result, the droplet is driven to move and accomplish other operations. The parallel-plate channel configuration has been also applied to dielectric liquids, although the principle is different from EWOD [2-4]. The liquid dielectrophoresis (L-DEP) is a force exerting on the entire nonconductive droplet to actuate it, while the electrohydro-force in EWOD is concentrated on the contact line. In a non-uniform electric field, the dipoles forming dielectric liquids tend to where the electric field intensity is larger. Creating, moving, cutting and merging of dielectric droplets have been successfully realized using square (or similar) type solid electrodes in the parallel-plate structure [5]. However, all these manipulations are effective only with fluids that are commonly squeezed between two plates with channel gaps less than 300 μm. As a result, the liquid volumes are confined to several microliters at most. In addition, such closed structures defy three dimensional (3D) control of droplet.

To solve these issues, the principle of dielectrowetting [6, 7] is applied in our device to achieve creating, transporting, splitting and merging of sessile droplets in open space. A recent report has showed that dielectrowetting, which evolves from liquid dielectrophoresis, produces superspreading (significant change in contact angle) and works for non-conductive fluids [8]. In the present device, the interdigitated electrodes generate fringing electric field penetrating into the dielectric liquid, and the corresponding L-DEP force changes the contact angle, even making the droplet to be a thin film. At the interface of liquid and air, L-DEP force P_{L-DEP} [9] is deduced to

$$P_{L-DEP} = \frac{1}{2}E^2(\varepsilon - 1)\varepsilon_0 \qquad (1)$$

where E is the electric field intensity, ε relative permittivity of fluid, and ε_0 permittivity of air. This equation shows that with increasing the electric field intensity and permittivity of dielectric liquid, the L-DEP force is elevated. Meanwhile, since the electric field generated by interdigitated electrodes decreases exponentially from the substrate surface, the L-DEP force varies with it, *i.e.*, decreases rapidly along the liquid-vapor interface from the three-phase contact line. This property makes dielectrowetting similar with EWOD in view of the force in the vicinity of contact line. As a result, the contact angle of dielectric droplet changes upon applying electrical potential. However, the difference from EWOD is that contact angle change is anisotropic in dielectrowetting. As the L-DEP force is in the direction of the interdigitated electrodes, the contact angle change is only observable perpendicular to the electrodes, while the contact angles and

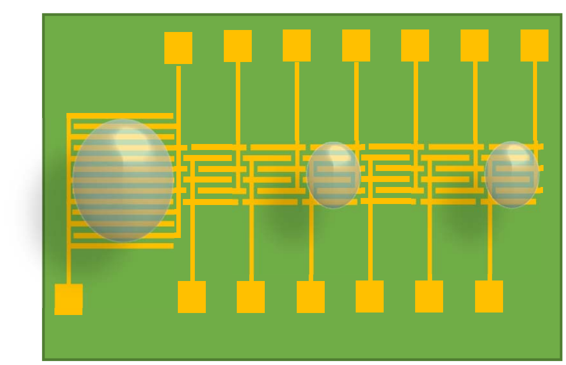

Figure 1: Schematic of experimental device (not in scale). The large electrode is 5.5 x 5.5 mm, and the small ones are 2 x 2 mm. The width and spacing of interdigitated legs are 50 μm.

boundaries along the electrodes remain the same. The large L-DEP force can spread a droplet to be a liquid film covering the energized electrodes, which makes it possible to digitize the fluid in open environment. Finally, controlling conductive liquids by dielectrowetting is also feasible in this study. To the author's knowledge, this dielectrowetting principle has not been developed for the above fundamental droplet manipulations (creating, transporting, splitting and merging).

EXPERIMENTAL

For the four droplet operations, we uniquely designed an array of individually addressable multiple electrodes on a glass wafer (Figure 1). Unlike electrowetting, each dielectrowetting electrode set consists of two coplanar interdigitated finger electrodes to generate a non-uniform electric field between them. For the sake of droplet moving smoothly from one electrode set to another, an interlocking pattern is placed in the spacing between two electrode sets. The overall electrodes consist of one large and six small electrode sets. The larger one (5.5 x 5.5 mm) is used for droplet spreading, as well as a reservoir to create new small droplets. The six small electrode sets (2 x 2 mm) are designed to attain discrete droplets, and complete the cutting, moving and merging. The width and spacing of the interdigitated electrode fingers are 50 µm. The patterned structures were fabricated by conventional photolithography and lift-off technique. The electrodes were made of 5 nm Cr and 200 nm Ag by E-beam evaporating. A 2-µm thick parylene C layer was deposited by a chemical vapor deposition (CVD) system (PDS 2010, Specialty Coating System) on top. Next, a thin Teflon-AF (DuPont, USA) was dip-coated and dried on a hotplate (55ºC, 10 min) to increase the initial contact angle.

Propylene carbonate and DI water with/without surfactant (TWEEN®20) were used for non-conductive and conductive working fluids, respectively. There are several advantages to choose propylene carbonate as the dielectric liquid. First, the high relative permittivity (ε = 65) enhances the L-DEP force according to the previous equation.

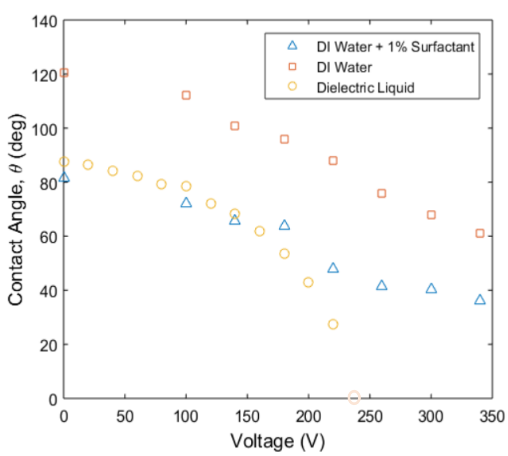

Figure 2: Contact angle vs voltage for three liquids. The frequency is 20 kHz for dielectric liquid, and 55 kHz for the other two liquids.

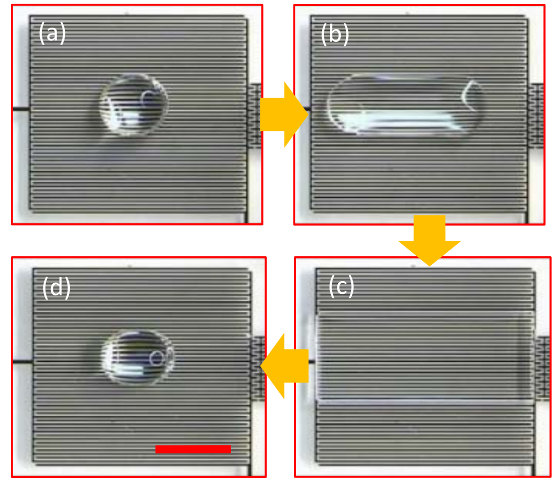

Figure 3: Top views of dielectric droplet spreading at 0 V, 220 V, 360 V and back to 0 V. The scale bar is 2 mm.

Secondly, the volatility of this liquid is very low, facilitating the operations in open environment. Thirdly, it is nontoxic and widely used as solvent. The surfactant was added to DI water with 1% volume rate and mixed thoroughly to decrease the contact angle of DI water droplet.

AC sinusoidal signal was provided by function generator (33220A, Agilent), followed by amplifying it using an amplifier (PZD 700, Trek). The voltages used were up to several hundred volts, and measured by oscilloscope (199 C, Fluke) in the root-mean-square mode. The CCD camera (CV S3200, JAI) with microscope lens recorded the real time motions that were saved in a computer.

RESULTS AND DISCUSSION

Droplet spreading

In dielectrowetting, the droplet response is highly sensitive to the frequency of AC voltage applied. It turns out that there exist some optimal frequencies at which the contact angles change most even under the same voltage. For propylene carbonate, such frequency is 20 kHz, while for DI water with and without surfactant, 55 kHz can be chosen as the working frequency. In figure 2, the contact angles of three liquids decrease as the voltage increases. For DI water, the contact angle changes from 120º (initial) to 61º (340 V voltage applied). However, the change range of EWOD is only 40º, after which contact angle is saturated. At the same voltage range, the contact angle of DI water with surfactant drops from 80º to 36º, with a change range of 44º. These results show that dielectrowetting is an effective method to modify the wetting property of conductive liquids. More interestingly, for dielectric liquid, complete wetting is achieved by providing sufficient voltage. The contact angle of propylene carbonate varies from 87.6º (0 V) to 0º (236 V). Above this value, the droplet becomes a thin liquid film and the contact angle is not measurable. Figure 3 shows how the droplet spreads by dielectrowetting. A 1.5-µL droplet is placed on the reservoir, with applying 220 V, 360 V, and finally 0 V. At 220 V, the droplet is elongated along the electrode fingers, without laterally crossing the boundaries. However, at 360 V, the droplet becomes a thin flat film, whose contact angle is 0º. In fact, the film will become thinner with larger voltages. After turning off the electrodes, the droplet is back to the semispherical shape. Therefore, dielectrowetting is a reversible process, which is important for droplet applications.

Droplet splitting and transporting

Dielectrowetting is a versatile method to control droplets, including splitting and transporting. In Figure 4, a small droplet (2.3 μL) is placed on the middle electrode set. Upon activating the three sets, it is elongated to form a thin liquid film, covering the activated area. Next, the middle electrode set is turned off and the droplet is cut into two individual droplets, which spread only on the second and fourth set. After powering off all the electrodes, the splitting process is completed, and two smaller hemispherical droplets are created. Similarly, the transporting of droplets is also accomplished by dielectrowetting, as shown from Figure 4(e) to (g). Although the split droplets are not contacting the neighboring electrode sets, the L-DEP force is strong enough to drive the droplets to cross the boundaries. Liquid films covering two electrodes are generated when the two electrode sets are on (Figure 4(e)). As the second and fourth electrodes are off, the droplets move to the ends of the electrode array. Finally, the two small droplets are located on the first and fifth sets after turning off all the electrodes.

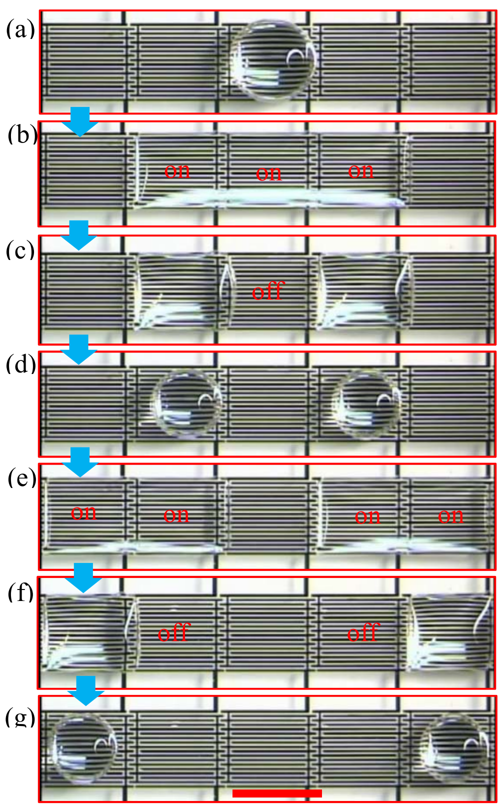

Figure 4: Sequential images of droplet splitting and transporting (dielectric fluid). The voltage is 360 V. The scale bar is 2 mm.

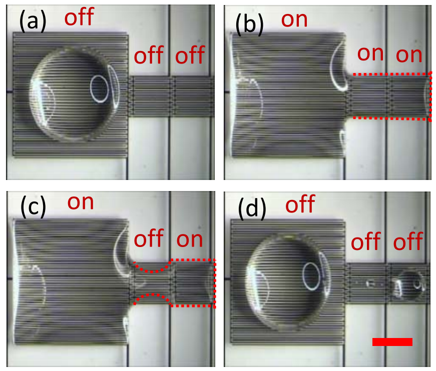

Figure 5: Four steps of droplet generating. The voltage is 360 V at 20 kHz. The scale bar is 2 mm.

Droplet creating

Figure 5 demonstrates the 4-step process of droplet generating. At the beginning, a 22-μL mother droplet is dropped on the reservoir. Next, the large electrode set and 2 small ones are turned on simultaneously. The droplet spreads to cover all the electrode area. It means that the L-DEP force is large enough to drive the droplet to cross the boundaries of electrode sets. The third step is to turn off the middle electrode set to segregate a small droplet from the mother droplet. From Figure 5(c), the neck is observed on the middle electrode set at the moment of turning off it. The liquid contracts due to the surface tension and eventually cuts the original droplet. Finally, all the electrodes are turned off and 0.9 μL new droplet is created. To maintain the minimum energy state, the droplet becomes a semispherical shape again after turning off all the electrodes.

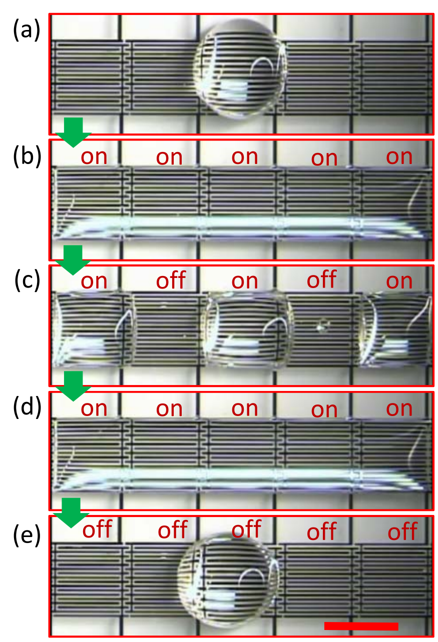

Figure 6: Splitting and merging of multiple (three) droplets in one operation for dielectric liquid (360 V). The scale bar is 2 mm.

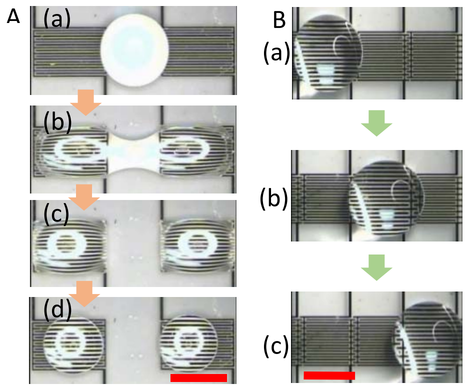

Figure 7: (A) Sequential images of DI water splitting. (B) Sequential images of DI water transporting. Scale bars are 2 mm. (55 kHz. 340 V)

Multi-splitting and merging

In addition to a single cutting, one droplet can be split into more than 2 daughter droplets at one operation, as shown in Figure 6. One 3.4-µL droplet is placed on the middle electrode set first. Then a long liquid film is obtained by turning on all the electrodes. After powering off the alternative electrode sets, 3 small droplets are created on the first, third and fifth electrode set. Such operation has not been reported for EWOD. Merging is performed by turning on all the electrodes and then off.

Aqueous droplet manipulations

To broaden the application of dielectrowetting, conductive liquids are utilized as well. At high frequency voltage, these liquids behave like dielectric fluid and dielectrowetting principle can be applied [10]. Figure 7(A) shows the splitting operation with DI water at 55 kHz, 340 V. One droplet is placed at the center area, followed by activating the two electrode sets simultaneously. Under the L-DEP force, a neck is formed in the middle and finally breaks up. Eventually, two individual water droplets with almost the same size are obtained. In Figure 7(B), transporting of DI water is also achieved by powering on the three electrode sets in turn. The volume effect on splitting is shown in Figure 8. At 55 kHz, 340 V, the threshold volumes are 4.5 µL, 5.8 µL and 8.2 µL for DI water with surfactant, propylene carbonate and DI water, respectively. Beyond these values, splitting is not realizable.

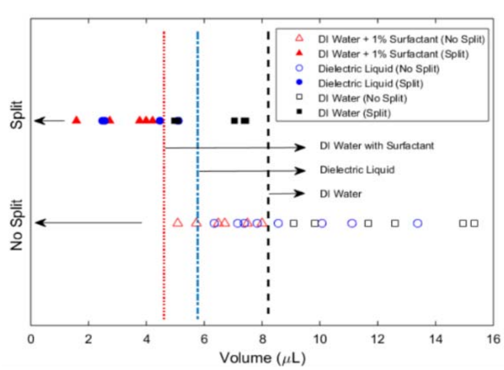

Figure 8: Volume effect on splitting (340 V, 55 kHz). The broken lines indicate threshold volumes where splitting occurs.

CONCLUSION

The paper presents a new method to manipulate droplets using dielectrowetting. Four fundamental operations, *i.e.*, creating, transporting, splitting and merging, are achieved successfully in open environment. The interdigitated electrode pattern is designed to change the wetting property of liquid on a single substrate. The contact angle of dielectric liquid decreases dramatically upon applying high voltage. The L-DEP force is large enough to deform droplet to be a thin liquid film. This mechanism is employed to build a digital microfluidic circuit to manipulate droplets versatilely. Not only dielectric liquid, but conductive droplet can also be split and moved by dielectrowetting.

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