Electrowetting on dielectric device with crescent electrodes for reliable and low-voltage droplet manipulation

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Digital microfluidics based on electrowetting on dielectric is an emerging popular technology that manipulates single droplets at the microliter or even the nanoliter level. It has the unique advantages of rapid response, low reagent consumption, and high integration and is mainly applied in the field of biochemical analysis. However, currently, this technology still has a few problems, such as high control voltage, low droplet velocity, and continuity in flow, limiting its application. In this paper, through theoretical analysis and numerical simulation, it is deduced that a drive electrode with a crescent configuration can reduce the driving voltage. The experimental results not only validate this deduction but also indicate that crescent electrode can improve the droplet motion continuity and the success in split rate.
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I. INTRODUCTION

In the last two decades, with the rapid development of microfabrication processes, the microfluidic technology has also made a major breakthrough, and digital microfluidics based on electrowetting on dielectric (EWOD) technology has become the new hot spot of microfluidics.¹ For droplet manipulations in digital microfluidics, voltage is applied to the drive electrode beneath a dielectric layer. This reduces the contact angle α , as shown in Fig. 1(a). When the amount of decrease in α is large enough, the droplets will move toward the direction of the actuated electrode. If voltages are applied to the drive electrode array according to certain spatiotemporal sequence, the droplets will move in a particular pattern. Fundamentals of EWOD include four basic manipulations of droplets: dispense, split, merger, and transport. In digital microfluidics, droplet manipulations are carried out on a flat plate and do not require micropumps, microvalves, micropipes, or other complex mechanical configurations, thus avoiding the contamination caused by the production, assembly, and cross-use of components with complex configurations. As a result, digital microfluidics has been applied more and more in lab-on-a-chip (LOC), which uses droplets as a carrier for cells, antibodies, proteins, enzymes, and other substances.^{2–5} In addition digital microfluidic technology has also been widely applied in optics,⁶ micro conveyor system,⁷ and other fields.

The magnitude of the voltage applied during droplet manipulation has a huge impact on the application prospect of digital microfluidics. This is because excessive voltage will form a strong electric field, not only causing irreversible damages to active substances contained in the droplets, such as cells, DNA⁸ and proteins, but also breaking down the dielectric layer and damaging the chip. In addition, under high voltage, the evaporation of the droplets on a microscale accelerates.⁹ To ensure the accuracy of the results of the biochemical reactions, a high velocity of the droplets on the chip is required, while the continuity of the droplet motion must be guaranteed. Hence, lowering the control voltage and ensuring the continuity of high-speed droplet motion on the chip are inevitable desirable trends in the field of digital microfluidics. Usually, the voltage is reduced via reducing

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FIG. 1. (a) The side view of the schematic diagram of a bipolar plate EWOD chip; (b) the top view of the EWOD force generated on the effective TCL for droplet transport; and (c) the top view of schematic diagram of three drive electrode structures.

contact angle hysteresis and friction by choosing silicone oil as the medium surrounding the droplet manipulation. Yet, the introduction of the silicone oil can cause pollution. In addition, Chang *et al.*¹⁰ used aluminum oxide and Lin *et al.*¹¹ used tantalum oxide to reduce the driving voltage with these materials of high dielectric constant. However, these methods involve complex processes, and the cost is high. Changa and Pak¹² used a twin-structure chip to increase the force on the droplets, reducing the driving voltage, but this method is not conducive to the assembly of the upper and lower plates of the chip. Banerjee *et al.*¹³ used strip-shaped electrodes to obtain high droplet speed. However, the width of this type of the drive electrodes is small, and a $1\ \mu\text{l}$ droplet can cover 2–4 actuated electrodes at the same time, increasing the difficulty in droplet manipulation.

Up to now, most of the previous researches on EWOD-driven electrodes of crescent configuration have been proposed,^{14,15} yet there has been no theoretical deduction or experimental tests regarding its performance in driving droplets. Here, based on the key fact that the force exerted on the droplet is proportional to the length of the cord formed by the effective three-phase contact line (TCL), theoretical analysis and numerical simulation are done. From this, it is deduced that crescent drive electrodes are not only optimal for reducing the driving voltage but also capable of improving the velocity and continuity of droplet motion. Meanwhile, from our experimental results, when the opposing layout is used, crescent drive electrodes can reduce the driving voltage required for droplet split by about 40%.

II. THE DESIGN OF THE DRIVE ELECTRODE PROTOTYPE AND NUMERICAL SIMULATION

As shown in Fig. 1(b), the circle denotes the contact circle of the droplet. The droplet is placed on top of the left and the right electrodes, and the right electrode is actuated.

The horizontal EWOD tension (f_{er}) generated at each point on the TCL induces the change of the apparent contact angle as shown in the following Lippmann–Young equation:^{12,16,17}

$$f_{er} = \frac{cV^2}{2}, \quad (1)$$

$$\gamma_{lv} \cos \theta_V = \gamma_{sv} - \gamma_{sl} + \frac{cV^2}{2}, \quad (2)$$

where c denotes the capacitance per unit area of the dielectric layer; γ_{lv} denotes the liquid-vapor surface tension; γ_{sl} denotes the solid-liquid surface tension; γ_{sv} denotes the solid-vapor surface tension; and V denotes the driving voltage. As can be seen in Fig. 1(b), the EWOD force (F_{ex}) acting on the droplet in the advancing direction (x) can be calculated by integrating the x -component of the horizontal EWOD tension (f_{er}) along the effective TCL on the actuated electrode^{12,16,17} as shown in the following equation:

$$F_{ex} = \int_{TCL} f_{er} \cos \phi dl = \frac{L}{2} cV^2, \quad (3)$$

where L denotes the length of the chord formed by the effective three-phase contact line and ϕ is as shown in Fig. 1(b). From Eq. (3), it can be seen that the EWOD force is proportional to the square of the driving voltage and proportional to the chord length L of the effective TCL on the actuated electrode. Under the same driving voltage, the larger the chord length L , the greater the EWOD force. Therefore, if the chord length L is relatively large, a relatively large EWOD force can be obtained, thereby achieving the goal of reducing the driving voltage in digital microfluidics.

Fig. 1(c) illustrates drive electrodes of square, jagged,^{18–20} and crescent shapes.^{14,15} When the outer contour of the electrode has the ratio $B/W \approx 1$, not only can the force exerted on the droplet be large but also can the difficulty of droplet manipulation be reduced. Hence, here the B/W ratio is set to be 1. The size of the outer contour of all three electrodes is 1×1 mm. With finite element analysis (COMSOL Multiphysics), numerical simulation of the force exerted on the droplet using the three shapes of drive electrode as shown in Fig. 1(b) was performed, and the relevant model parameters used in numerical simulation are illustrated in Table I. When performing the numerical simulation, the droplet can be approximated as a dielectric.^{19,21,22} According to the law of conservation of charge and Laplace's equation

TABLE I. Parameters used in numerical simulation.

Parameter	Value
Electrode dimensions ($L \times L$)	$1.4 \times 1.4 \text{ mm}^2$
Electrode spacing (g)	$20 \text{ }\mu\text{m}$
Dielectric layer thickness	$1 \text{ }\mu\text{m}$
Droplet volume	$1 \text{ }\mu\text{l}$
Droplet radius	$702 \text{ }\mu\text{m}$
Droplet base radius	$680 \text{ }\mu\text{m}$
Static contact angle	118°
Droplet conductivity	$5.5 \times 10^{-6} \text{ S/m}$
Relative permittivity of droplet	80
Relative permittivity of dielectric	3.2
Relative permittivity of air	1
Actuation voltage (V)	50 Vrms
Plate gap (h)	$300 \text{ }\mu\text{m}$

$$\nabla(\sigma \nabla V) = 0, \quad (4)$$

$$\nabla(\varepsilon \nabla V) = 0, \quad (5)$$

where $\varepsilon = \varepsilon_r \varepsilon_0$, ε_r is the relative permittivity of the medium, ε_0 is the permittivity of free space, σ is the electrical conductivity of the droplet, and V is the applied voltage which generates an electrostatic field in the vicinity of the droplet. The electric force on the droplet caused by this electric field can be expressed using Maxwell stress tensor, according to

$$F_i = \iint_{\Sigma} \sum_{j=1}^3 T_{ij} ds_j, \quad (6)$$

$$T_{ij} = \varepsilon \left(E_i E_j - \frac{1}{2} \delta_{ij} E^2 \right), \quad (7)$$

where F_{ij} is the EWOD force component in rectangular coordinate system and T_{ij} is the Maxwell stress tensor. Actuation forces were calculated over a series of droplet locations along its actuation path. Since the integration of the Maxwell stress tensor is dependent on mesh geometry and density, a uniform swept mesh on droplet surfaces is used to allow for a fine mesh on the droplet leading and trailing faces without considerably increasing the total number of elements in Fig. 2(a).

Fig. 1(b), (A) and (B), illustrates a droplet with a fixed volume partially overlapping two adjacent control electrodes in a digital microfluidics system. A droplet with a circular footprint is centered at a distance of x from the center of electrode 1. As can be seen here, as the droplet travels away from electrode 1 (left electrode), the EWOD forces exerted on the droplet moving from the inactive electrode to the actuated electrode as shown in Fig. 1(b) are labeled as crescent-right (Fig. 2(b)). Conversely, the other direction of droplet motion as shown in Fig. 1(b) is labeled as crescent-left.

Using actuated electrode of crescent shape, when the droplet moves to $600 \mu\text{m}$ ($x = 600 \mu\text{m}$), the force has already reached the maximum value of $4.2 \mu\text{N}$ (Fig. 2(b)). Yet, when actuated electrode of square or jagged shape is used, the droplet needs to move the distance of an entire drive electrode to reach the maximum force. Conversely, when the left electrode in Fig. 1(b) is actuated and the droplet moves toward the left direction, the force on the droplet moving on a square or a jagged electrode is not affected, and the force on the droplet moving on a crescent electrode also reaches maximum at a moving distance of $600 \mu\text{m}$. Fig. 2(b) and Eq. (3) show that the moving distance required to reach the maximum force is shortest when crescent electrode is used. Based on this analysis, this means that under the same condition over a short distance, the force on the droplet is the largest with crescent electrode.

Figs. 3(a)–3(d) show the schematic diagram of four types of drive electrode layouts for droplet split. In every electrode set, the left and the right electrodes are actuated, and the middle electrode is inactive. The widths of the four layouts are the same. As soon as voltages are applied to the two sides, the droplet starts elongating longitudinally. Then, more and more liquid accumulates over the actuated electrodes along with the driving voltage increasing while the amount of liquid on the center electrode reduces to keep the total droplet volume constant. Finally, only when the combination of material properties, configuration parameters, and applied voltages is such that the radii of curvature of the neck continue to decrease, the splitting of the droplet can be completed.

In order to understand the interrelation between the parameters and the condition necessary for successful splitting, there is a static analysis performed by Cho *et al.*^{23–25} Considering Laplace's equation for pressure difference across the liquid-air interface and the Lippmann-Young's relation between the contact angle and voltage, the following relation was derived:

$$\frac{R_2}{R_1} = 1 - \frac{R_2}{h} \frac{\varepsilon_0 \varepsilon_r}{2\gamma_{lv}} V^2, \quad (8)$$

FIG. 2. (a) A droplet three-dimensional simulation model and the uniform swept mesh on droplet surface. (b) Comparison of numerical simulation results of EWOD forces exerted on the droplet with different drive electrode structures.

where R_1 and R_2 are the respective radii of curvature of the neck and the front of a droplet along the both sides as shown in Fig. 3(a), h is the inter-plate spacing between upper and lower plates, and t and ϵ_r are the combined thickness and equivalent dielectric constant of the dielectric and hydrophobic coating, respectively. Equation (8) can be used to understand the key parameters and their interrelation that controls the droplet split process.

Figs. 3(a), 3(b), 3(c), and 3(d) illustrate the square, jagged, crescent, and opposing layouts of crescent electrodes, respectively. The relevant parameters for the numerical simulation are shown in Table I. Fig. 3(e) shows the results of the numerical simulation of the force on the droplet in droplet split using four different electrode layouts. Driving voltage is the only parameter that changes in the simulation, whereas other parameters remain the same.

As can be seen, in general, the force increases as the driving voltage increases. When the driving voltage is 50 V, the force on the droplet on crescent electrode with an opposing layout reaches the maximum value of $8.5 \mu\text{N}$; when the driving voltage increases to 60 V, the force is approximately zero, suggesting that the droplet had been successfully split. In contrast, the

FIG. 3. (a)–(d) The schematic diagrams of droplet split using the four different drive electrode layouts. (e) Comparison of numerical simulation results of EWOD forces exerted on the droplet on four different electrode layouts.

force on the droplet on square, jagged electrode, sequential layout reaches the maximum value of $8.5 \mu\text{N}$ when the driving voltage is 90 V, 70 V, and 90 V, respectively, as shown in Fig. 3(e). Similarly, the force is zero when the driving voltage is to go on with increasing, this means that these three layouts can also split a droplet successfully; nevertheless, higher driving voltage is needed. An important conclusion that can be determined from these data is that the driving voltage needed for successful droplet split is lowest when an opposing layout of the crescent electrodes is used. This layout is conducive to droplet split.

III. EXPERIMENTAL TESTS

The proposed device as shown in Fig. 1 consists of 1×5 array of control electrodes of $1.4 \times 1.4 \text{ mm}^2$ ($B = W$) with $20 \mu\text{m}$ electrode spacing (g). As far as the radius of the curve in crescent electrode configuration is $750 \mu\text{m}$, the dimension of jaggies in jagged electrode is $0.45 \times 0.21 \text{ mm}^2$. The devices were prepared by the following processes. The ITO-coated^{26–28} glass (Kaiwei electronic devices Limited, Zhuhai, China) plates were rinsed by acetone and de-ionized (DI) water to remove contaminants before the 120°C dehydration bake. Electrodes were patterned using conventional lithographic techniques. Briefly, AZ 1512HS positive photo-resist (Micro Chemicals, Ulm, Germany) was spin-coated onto the substrates (500 rpm for 10 s and 3000 rpm for 30 s), which were then soft-baked at 95°C for 3 min. Coated substrates were

exposed to UV radiation (70 mW/cm^2) for 20 s through a bright-field mask (SVG Optronics, Co., Ltd., Suzhou, China) designed in AUTOCAD2007 (Autodesk, San Rafael, CA). The pattern was then developed using a 3:8 solution of AZ 400 K (MicroChemicals, Ulm, Germany) in DI water. After dehydrating at 90°C for 4 min, the ITO was etched in a 4:2:1 (v/v/v) solution of HCl, H_2SO_4 , and HNO_3 for 21 s. The remaining photoresist was then removed with acetone.

Dielectric layer deposition was achieved by spin-coating SU8-3 (MicroChemicals, Ulm, Germany) onto the substrate with the patterned electrodes (500 rpm for 10 s and 3000 rpm for 40 s). Two 180 s post-bakes were necessary at 65°C and 95°C , respectively. After removing the SU8 from the contact pads with acetone, the layer was exposed to UV light (70 mW/cm^2) for 25 s. Finally, devices were baked at 95°C for 3 min and at 160°C for 0.5 h.

Hydrophobic layer deposition was performed on both plates of the device by spincoating Teflon-AF1600 (DuPont, Wilmington, USA) onto the devices (500 rpm for 5 s and 1000 rpm for 60 s). Devices were then baked at 160°C for 10 min. The final devices had layers with a thickness of $2 \mu\text{m}$ and 50 nm for SU8 and Teflon, respectively. During the experiment, different layers of double-sided adhesive tape were used to secure the upper and lower plates of the chip. The upper plate is connected to the null electrodes. The drive electrodes were connected to a sinusoidal signal of frequency 100 Hz and magnified through a voltage amplifier circuit. Different volume of deionized water droplets was used, and medium for droplet manipulation was air in all experiments. Motion was then recorded using a CCD camera (Basler acA1300-30gc, Exton, PA) connected to a motorized zoom lens (Navitar 12X Body Tubes, Rochester, NY).

FIG. 4. Video frame images of the motion of a $1 \mu\text{l}$ droplet on chip with crescent drive electrodes (the inter-plate spacing h is $300 \mu\text{m}$ and the scaling bar is 1 mm).

Because ITO glass has good transparency, yet turns opaque after coated with the SU-8 photoresist layer, in the video frame images of droplet motion the contours of the drive electrodes were outlined with dashed boxes, as shown in Figs. 4(a) and 6(a). The droplet moves from left to right in the electrode array shown in Fig. 4(a). It is obvious that in Figs. 4(b)–4(d), the droplet movement is smeared. The presence of this smear indicates high-velocity of the droplet motion. The frame rate of CCD camera used in our experiment is 50 frames per second. A few of frames of droplet motion were chosen to demonstrate the characterization of droplet movement, as shown in Fig. 4. The droplet presents a fast-moving state in some specific frames due to the high droplet velocity. We speculate that the frame rate and the high droplet velocity are mismatched in some specific frames. That is to say that the droplet shape is fuzzy and smeared in some frames (Figs. 4(b)–4(d)).

FIG. 5. (a) Comparison of average velocities of a $1\ \mu\text{l}$ droplet at different driving voltages. (b) Comparison of the number of successful movements of a $1\ \mu\text{l}$ droplet in 100 movement cycles.

A comparison of a $1\ \mu\text{l}$ droplet average speed on chips using three types of drive electrode (four types of array as mentioned above) at the same inter-plate spacing $300\ \mu\text{m}$ and different driving voltages is furnished in Fig. 5(a). From Fig. 5(a), it is clearly shown that the current chip with crescent electrode reported here provides very high droplet velocity at a relatively lesser actuation voltage compared with other two types of drive electrode. At voltage 20 V, the droplet on a chip with crescent electrode can be successfully driven to motion, and the average speed¹³ comes out around 5 mm/s. As the driving voltage increases, the average speed of the droplet on the chip with crescent electrodes remains highest among the three electrode structures. Calculation reveals that the average speed of the droplet in air medium is found to be around 120 mm/s (Refs. 13 and 29) when the driving voltage is 40 V; in contrast, under the same experimental conditions, the $1\ \mu\text{l}$ droplet average speeds on the other two types of drive electrode are both substantially smaller than 120 mm/s, as illustrated in Fig. 5(a). In addition, Fig. 5(a) also shows that the average speed of the droplet moving from left to right and that from right to left on the crescent electrode array as shown in Fig. 4(a) is approximately equal. Note here that the effective TCL plays an important role in EWOD force, and the left crescent electrode configuration enables a rather larger effective TCL like jagged electrode structure^{18–20} when droplet moves from the right electrode to the left, namely, crescent-left as shown in Figs. 2(b) and 5. Under the same driving voltage, such a larger effective TCL will make droplet obtain a larger driving force compared with the square and jagged electrodes. Therefore, it interprets well that the droplet can be able to obtain a similar average speed on the chip with crescent electrodes no matter what the droplet moves from left to right or that from right to left as shown in Figs. 2(a) and 4(a).

Under the same driving voltage, the crescent electrode configuration can always ensure the continuity of the droplet movement. Yet with the other electrode configurations, sometimes the droplet cannot be successfully driven. In this case, the drive electrodes need to be actuated repeatedly to ensure a continuous movement of the droplet. To test the continuity of the droplet movements driven by the three types of electrodes, herein a movement cycle is defined as the process of the droplet completing the movement from left to right or from right to left across the five electrodes in the array as shown in Fig. 4(a). Fig. 5(b) shows the number of successful movement cycles without repeat actuation of the drive electrodes out of 100 attempts at different driving voltages, using different electrode configurations, but otherwise under the same experimental conditions. Obviously, the continuity is the highest when the crescent electrodes are used among the three electrode configurations. This is mainly because the shape of the crescent electrodes in the horizontal direction is of regular semicircular shape, and its distance to the circular cross-section of the droplet is rather uniform. As a result, the force on the droplet is evenly distributed, thereby ensuring the continuity of the droplet movement.

FIG. 6. Video frame images of the split of a $1\ \mu\text{l}$ droplet (the inter-plate spacing $h = 100\ \mu\text{m}$).

Fig. 6 shows the video frame images of splitting a $1\ \mu\text{l}$ droplet using crescent drive electrodes with an opposing layout as illustrated in Fig. 3(d). Fig. 7(a) shows the relationship between the minimum applied voltage and the droplet volume under the same experimental conditions using four different electrode layouts. As can be seen in Fig. 7(a), the driving voltage required for droplet split increases as the droplet volume increases. Yet among the four electrode layouts, the driving voltages needed with an opposing layout of the crescent electrodes remain to be the smallest. In addition, when the droplet volume is $2\ \mu\text{l}$, the minimum voltages required with the other three electrode layouts are nearly 100 V. Even though this can split the droplet, due to the high voltage, the dielectric layer of the EWOD chip is broken down. In comparison,

FIG. 7. (a) Comparison of the minimum driving voltages required for successful split of droplets of different volumes (the inter-plate spacing $h = 100\ \mu\text{m}$). (b) Comparison of the number of successful droplet splits using electrodes of different shapes ($V = 50\ \text{V}$).

with the opposing electrode layout, the required voltage is 75 V, and no dielectric breakdown occurs. This result fully demonstrates that crescent electrodes in an opposing layout can reduce the driving voltage needed for droplet split, by about 40%.

Fig. 7(b) shows the number of successful droplet splits using different electrode layouts. In order to eliminate the impact of errors in droplet evaporation and volume, for each layout, the droplet split experiments were conducted 10 times, and the number of successful split was compared. In the experiments shown in Fig. 7(b), the driving voltage is fixed at 50 V, and the volumes of the deionized water droplet are $1\ \mu\text{l}$. When the inter-plate spacing is $50\ \mu\text{m}$ or $100\ \mu\text{m}$, the numbers of successful droplet split using the four electrode layouts (opposing, sequential, jagged, and square) are the same. But when the inter-plate spacing $h \geq 150\ \mu\text{m}$, for the same spacing, the number of successful droplet splits is the highest when crescent electrodes of an opposing layout are used, and with all the other three layouts, the number of successful splits is largely the same.

Fig. 8 portrays the schematic of droplet split of three types of droplet volume using four types of drive electrode layouts as shown in Fig. 3. As discussed above, under the same experimental conditions, four types of drive electrode layouts for droplet split show different performance. Out of the four layouts, the opposing layout of crescent electrodes is optimal for droplet split. It is noted here that number 1 dotted circle represents a certain volume of droplet whose two sides overlap the actuated electrodes entirely, as shown in Fig. 8. Similarly, number 2 dotted circle represents a certain volume of droplet whose two sides overlap the actuated electrodes partially. In contrast, number 3 dotted circle represents a certain volume of droplet which has no contact with the actuated electrodes. Using four types of drive electrode layouts, there is the possibility that number 1 droplet could be split successfully if only the driving voltage is larger enough or the plate gap h is sufficient small (as shown in Fig. 7). Using any given electrode layout, the successful droplet split will not happen for the number 3 droplet by reason of no EWOD force exerted on the droplet. In the case of number 2 droplet, using square, jagged, and sequential electrode layout, the elongating EWOD force from two sides might be smaller or unequal compared with the opposing layout, this is because the opposing layout of crescent electrodes conforms with the droplet three-phase contact line (Fig. 8), this will facilitate larger EWOD forces from the beginning of motion, even though it will not increase forces above its maximum. Therefore, a rather larger composite EWOD force exerted on droplet in opposing crescent electrodes layout can be obtained at the same driving voltage. To sum up, based on the above numerical simulation and the experimental results, we conclude that an opposing layout of the crescent electrodes is conducive to droplet split.

FIG. 8. Schematic diagrams of droplet split of three types of droplet volume. (a)–(d) Square, jagged, sequential, and opposing drive electrode layouts.

IV. CONCLUSION

In summary, the results of the present study reveal that compared to square and jagged electrodes, crescent electrodes can substantially decrease the applied voltage for driving droplets, achieving the highest average speed as well as the highest continuity of the droplet movement in air-filled environment. In addition, when an opposing layout of the crescent drive electrodes is used, the driving voltage is lowest, and the success rate of droplet split is the highest among four different electrode layouts. Most importantly, experimental results agreed with the theoretical deduction and the numerical simulation results, which suggest utility in device design and fabrication. Therefore, it can be concluded that the crescent drive electrodes in digital microfluidics are conducive to the continuous transport and the split of droplets for integrating the device with others.

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