A novel actuation method of transporting droplets by using electrical charging of droplet in a dielectric fluid

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We evaluate the feasibility of manipulating droplets in two dimensions by exploiting Coulombic forces acting on conductive droplets immersed in a dielectric fluid. When a droplet suspended in an immiscible fluid is located near an electrode under a dc voltage, the droplet can be charged by direct contact, by charge transfer along an electrically conducting path, or by both mechanisms. This process is called electrical charging of droplet (ECOD). This charged droplet may then be transported rapidly by exploiting Coulombic forces. We experimentally demonstrate electrical actuation of a charged droplet by applying voltage sequences. A charged droplet is two dimensionally actuated by following the direction of the electrical field signal. The droplet does not contact the surface of the microfluidic chip when it moves. This characteristic is very advantageous because treatments of the substrate surfaces of microfluidic chip become simpler. In order to test the feasibility of using ECOD in a droplet-based microreactor, electrocoalescence of two oppositely charged droplets is also studied. When two droplets approach each other due to Coulombic attraction, a liquid bridge is formed between them. We postulate that if the applied electric field is weaker than a certain critical level, the two droplets coalesce instantaneously when the charges are exchanged and redistributed through this liquid bridge. © *2009 American Institute of Physics.*
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I. INTRODUCTION

The capacity to use electricity to control the shape and position of droplets on solid surfaces has led to a dynamic new field of research.[1] The behavior of electrically driven droplets on a substrate under an electric field has potential applications in various areas, including biomicroelectromechanical systems (bio-MEMSs).[2,3]

A charged droplet can be used as an individual microreactor in bio-MEMS.[4] Many reported that micro-total-analysis system designs consist of a chemical analysis system microfabricated and integrated on a substrate, but there are some drawbacks in the continuous-flow systems such as surface contamination and dead volume. These problems could be solved using discrete flow (i.e., droplet-based) manipulation, which has several advantages over the traditional continuous-flow microfluidic systems. In particular, precise manipulation of droplets in microfluidic devices has allowed development of high-throughput reactors that use minute quantities of reagents. Currently, the main application of droplet-based microfluidics is electronically controllable transportation of droplets.

We exploit the phenomenon that electrical charging of a water droplet occurs on a bare electrode immersed in silicone oil. As the droplet approaches the bare electrode, it is deformed into an elongated shape (Fig. 1). Since the water droplet is a better conductor than the dielectric medium, it is more easily polarized than the oil. So, the charges opposite to the electrode accu-

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FIG. 1. Moment of contact: The drop can be charged by direct contact, by transfer of charge along an electrically conducting path, or by both mechanisms.

mulate at the water/oil interface near the electrode. Further, because the electrode has a planar shape, more charges accumulate at the interface near the pole of the droplet facing the electrode. The electric field strength increases with the accumulated charges and the decreasing distance between the droplet and the electrode. Thus, a droplet is deformed to a shape with a sharp tip as it approaches the electrode.[5,6] This sharp tip eventually makes a contact to the electrode and the droplet acquires charges from the electrode through it. This phenomenon is called electrical charging of droplet (ECOD).

In ECOD, one important challenge is to determine how much charge a droplet can acquire at the electrode. Recently, electrical charging of a deformable droplet by ECOD was experimentally studied by Jung *et al.*[7] This provides the basic information for the present work as ECOD-based microfluidics.

In this work, we evaluate whether a droplet charged by using ECOD can be manipulated individually based on the results of previous researches. If such manipulation is possible, droplets may be used as microreactors in which molecules, cells, or particles are embedded. To test the feasibility, we observe also the electrocoalescence of two oppositely charged droplets. By combining the electric charge distribution of a droplet and the applied electric field on the device, fine control over individual droplets may be achieved to design an ECOD-based microfluidic system.

II. DIGITAL MICROFLUIDICS: ECOD PRINCIPLE

Digital microfluidics (DMF) is a method of handling liquids in droplet forms. Because discrete droplets can be handled individually (i.e., digitized), ECOD can be regarded as a form of DMF. DMF has the advantages that droplet transport is relatively fast and results in relatively little surface contamination. Therefore, we have focused on ECOD in immiscible fluids.

A. ECOD detection area

We construct a platform for handling conducting droplets by using electrode dots on a chip substrate (Fig. 2). To configure the microcontroller system, a programmable multichannel high voltage supply is connected through an electrical switch to the electrodes arrayed on a hydrophobic Teflon plate. Water forms spherical droplets on a hydrophobic surface; these are suitable for ECOD actuation. Droplet actuation is achieved sequentially by applying voltages $(a \rightarrow b)$ to bare copper electrodes. The sequential voltages we used are a three-phase profile generated by using LAB-VIEW™: One voltage sequence such as $[+:-:0]$ was first evaluated, and then voltage sequence $[0:+:-]$ is evaluated, which is proved to be the time sequence for droplet transport. When a voltage is applied to the electrodes, the droplet approaches the electrode. The droplet acquires the same charge as the electrode and is then repelled toward the opposite electrode. At the moment that the droplet arrives at the opposite electrode, the voltage of the previous electrode pair is disconnected and the voltage is simultaneously supplied to the next electrode pair. The recharged droplet is also translated according to the electric field signal.
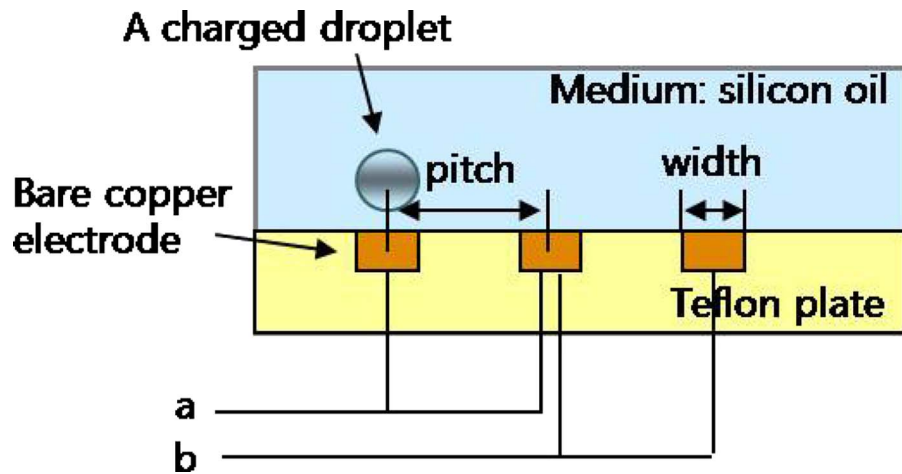
FIG. 2. The schematic view of the concept of a sequential signal and ECOD contact area.

B. Microcontroller system and sequential voltages

We construct a microcontroller system that applies a sequential voltage signal to control the motion of droplets (Fig. 3). A programmable multichannel high voltage supply is connected to the components in relays. These pins are connected to a Ni terminal block and use Lab-view™ as an electrical switch; the pins work at 12 V dc nominal voltage. When the 12 V dc signal is switched through the LAB-VIEW™ program in the intended direction, an electric field is supplied to the corresponding electrodes.
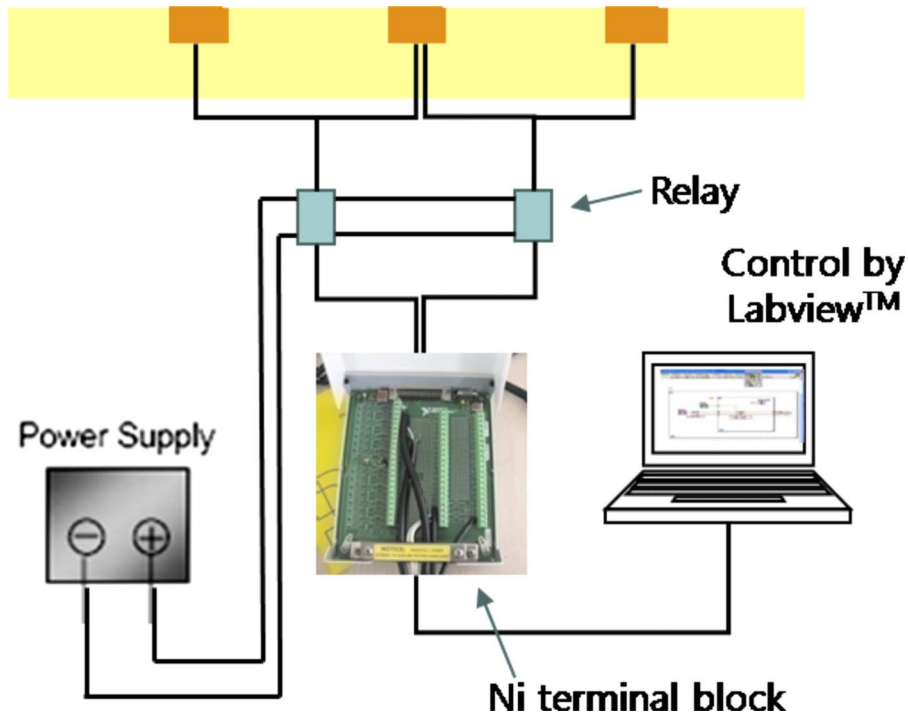

FIG. 3. The concept of the custom-made microcontroller system: The droplet is manipulated on an electrode matrix according to the electric field signal.
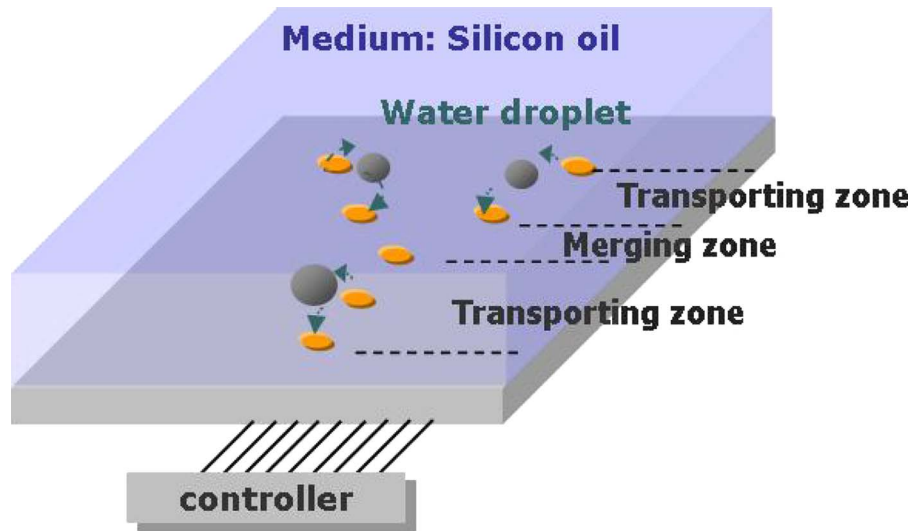
FIG. 4. The concept of the device by using electrode dots on the hydrophobic Teflon surface: The pitch (distance between the centers of electrodes) is ~10 mm and the diameter of electrode is ~1.8 mm.

III. MATERIALS AND EXPERIMENTAL CONDITIONS

In the experiment, we demonstrate the two-dimensional transport of a single de-ionized (DI) water droplet according to the voltage sequences. We also observe the coalescence of two oppositely charged droplets.

A. Device with electrode dots

In the device with electrode dots (Fig. 4) a Teflon plate is used as a hydrophobic surface. A circular noninsulated copper electrode is used, which has a diameter of ~1.8 mm. The distance between the centers of the electrodes is ~10 mm. These electrodes are connected to the droplet-microcontroller system. The electrode dot matrix consists of 3×3 units and its area is 40×40 mm^2.

B. Experimental setup

In the experimental setup (Fig. 5), a transparent rectangular acrylic test cell (internal dimensions $40 \times 60 \times 45$ mm^3 $[L \times W \times H]$) was placed between a light source and a microscope. Bare copper electrodes are connected to a dc voltage power supply (Trek, Inc.). To observe translation and coalescence of droplets, a Cannon digital camera (125 frames/s) and a Photron Fastcam 1024 PCI Model 100 K (3000 frames/s) are mounted on the front and upper side of the test cell, respectively. Relevant images are extracted using image analysis. A single DI water droplet ($\varepsilon = 77.75\varepsilon_0$, where $\varepsilon_0 = 8.854 \times 10^{-12}$ F m^{-1}, $\sigma_w = 1.2 \times 10^{-4}$ S m^{-1}, $\mu_w = 8.90 \times 10^{-4}$ kg m^{-1} s^{-1}, and $\rho_w = 997.75$ kg m^{-3}) is used as the conducting droplet and a silicone oil (KF-96 series, Shin-Etsu Silicone, $\varepsilon = 2.75\varepsilon_0$, $\sigma \leq 10^{-13}$ S m^{-1}, $\rho = 957.24$ kg m^{-3}) with kinematic viscosities of 10 cS (centistoke: 1 cS$= 10^{-6}$ m^2 s^{-1}) was used as the dielectric fluid medium. The corresponding drop volume is 2 μL. We apply external voltages of 1 kV to the electrodes set 10 mm apart. An Eppendorff micropipette is used to inject microsized water droplets into the cell.

C. Experimental procedure

We prepare the device with electrode dots and silicone oil fluid in the test cell. Then we put a water droplet near an electrode using a micropipette and apply a dc electric field to the electrodes. The experiment is completed as quickly as possible after the silicone oil is added to the test cell. Caution is also taken to preclude change in the properties of the silicon oil due to high electric
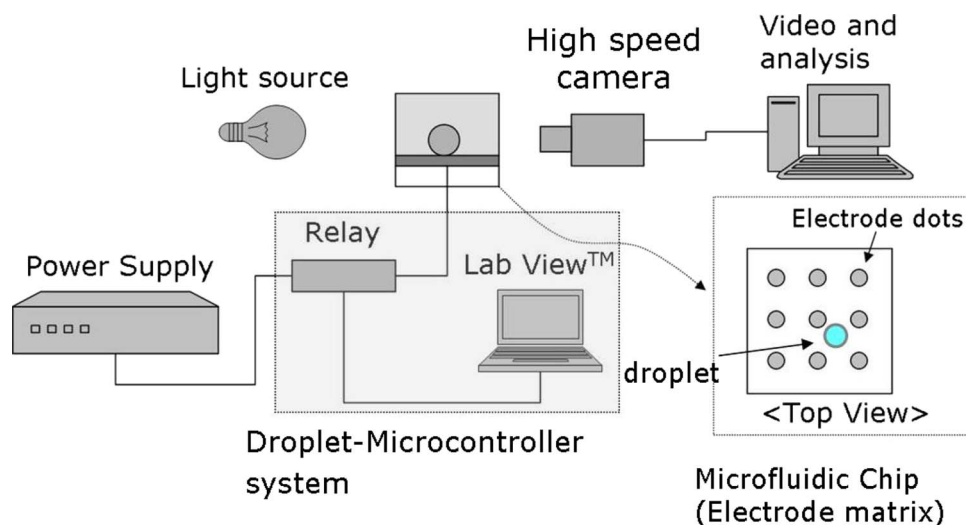
FIG. 5. Schematic view of experimental setup containing the droplet-microcontroller system.

field. The control of translating motion of a droplet is as follows. At the moment the droplet approaches the electrode, we cut off the voltage at the first two electrodes and supply the sequential voltage simultaneously to the nearest two electrodes in the intended direction.

The electrocoalescence of two droplets can be achieved in the same manner. We put two droplets individually near the electrodes using a micropipette, and then a dc voltage is supplied to the electrodes. To help visualize the electrocoalescence, we use an ink solution (50 vol %). The high speed camera is used for detailed observation.

IV. RESULTS

In the experiment, a droplet was actuated two dimensionally along the electric field signal. Detailed observation of the moment of contact was achieved. We also performed the feasibility test of electrocoalescence of two droplets as an essential step for being used as droplet-based microreactors. A close-up view of the moment of electrocoalescence was also achieved.

A. Electrical actuation of a microsized droplet on the substrate: Straight and turning motion

An initially uncharged conducting water droplet suspended in silicon oil starts to move slowly due to dielectrophoretic force when a dc electric field is applied.[5] In the present work, however, the droplet was pushed manually to the electrode to make an initial contact in a short time. After the initial contact, the droplet becomes charged by ECOD and starts to hop between electrodes along the electric field line. The silicon oil ($\rho = 957.24$ kg/m^3) has almost the same density as water ($\rho_w = 997.75$ kg/m^3) and the electric field line forms an arc between the electrodes.

So, the motion of a droplet was controlled easily by the sequential electrical signal (Fig. 6). For straight motion [Fig. 6(a)], we applied the electric field between the third and sixth electrodes and then we applied the electric field between the sixth and ninth electrodes immediately after the voltage between the third and sixth electrodes was switched off. The droplet charged by the third electrode passed to the sixth electrode and ultimately arrived at the ninth electrode. After the droplet acquired a negative charge from third electrode it began to move toward the sixth electrode, which had the opposite charge. When the droplet arrived at the sixth electrode, the voltage between the third and sixth electrodes was switched off at the same time that the voltage between the sixth and ninth electrodes was switched on. The droplet discharged its negative charges and simultaneously acquired positive charges from the positively charged sixth electrode. The recharged droplet then moved to the next (ninth) electrode due to Coulombic force.
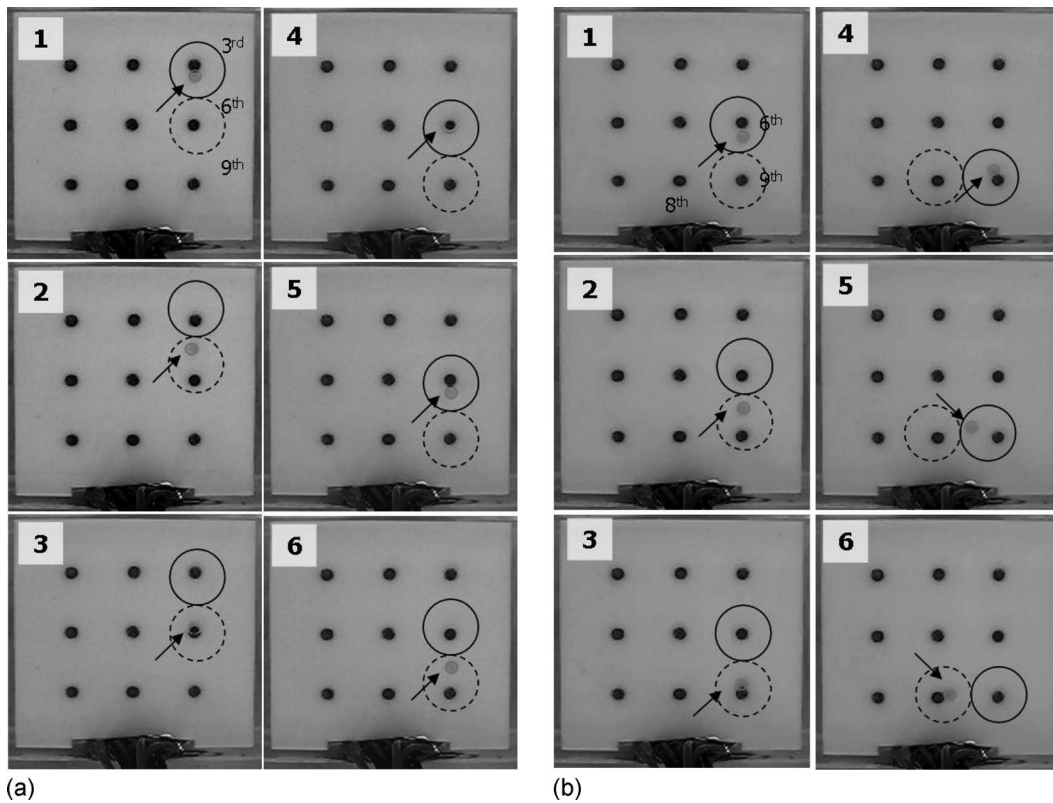
FIG. 6. The sequential images of the translated droplet. (a) The droplet charged by the third electrode passed to the sixth electrode and ultimately arrived at the ninth electrode (straight motion). (b) The droplet charged by the sixth electrode bypassed the ninth electrode and traveled to the eighth electrode (right turn motion). (Solid circle, positive electrode; dotted circle, negative electrode.)

Similarly, for the turning motion, we applied the electric field between the sixth and ninth electrodes and then applied the electric field to the eighth and ninth electrodes at the same time that the voltage between the sixth and ninth electrodes was switched off [Fig. 6(b)]. The droplet charged by the sixth electrode bypassed the ninth electrode and traveled to the eighth electrode.

A side view of a translating droplet [Fig. 7(a) (enhanced)] shows that the droplet does not contact the surface of substrate when it moves; instead it "hops" over the surface [Fig. 7(b)]. Since the density difference is small, a droplet moves along the arc over the surface formed by the electric field. So, a droplet hardly contacts the substrate when it moves. Now, the Bond number can also be estimated. The Bond number is a measure of the importance of the body force relative to the surface tension force and it is defined as

$$\mathrm{Bo} = \frac{\Delta \rho g l^2}{\gamma},$$

where $\Delta \rho$ is the density difference, g is the gravitational acceleration, l is the characteristic length of the droplet (here the diameter of undeformed droplet), and γ is the interfacial tension. For the present case, the Bond number is about 0.03. This small value indicates that the surface tension force dominates and a stationary droplet remains spherical. The contact area between the droplet and the substrate, therefore, is minimized in the form of the tip of the droplet. In ECOD, therefore, a substrate with a moderate hydrophobicity, such as Teflon plate, is enough for manipulating the droplets. Critical surface treatment of the chip substrate, such as thin hydrophobic coating and careful chemical coating, is not needed.
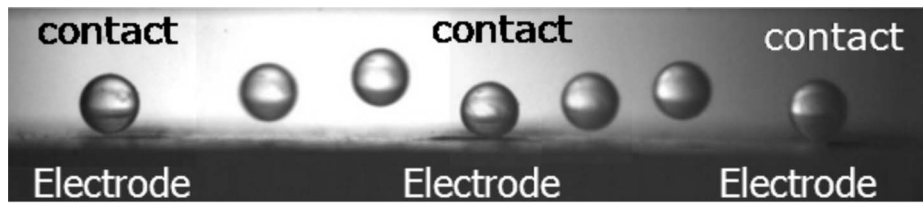
(a)
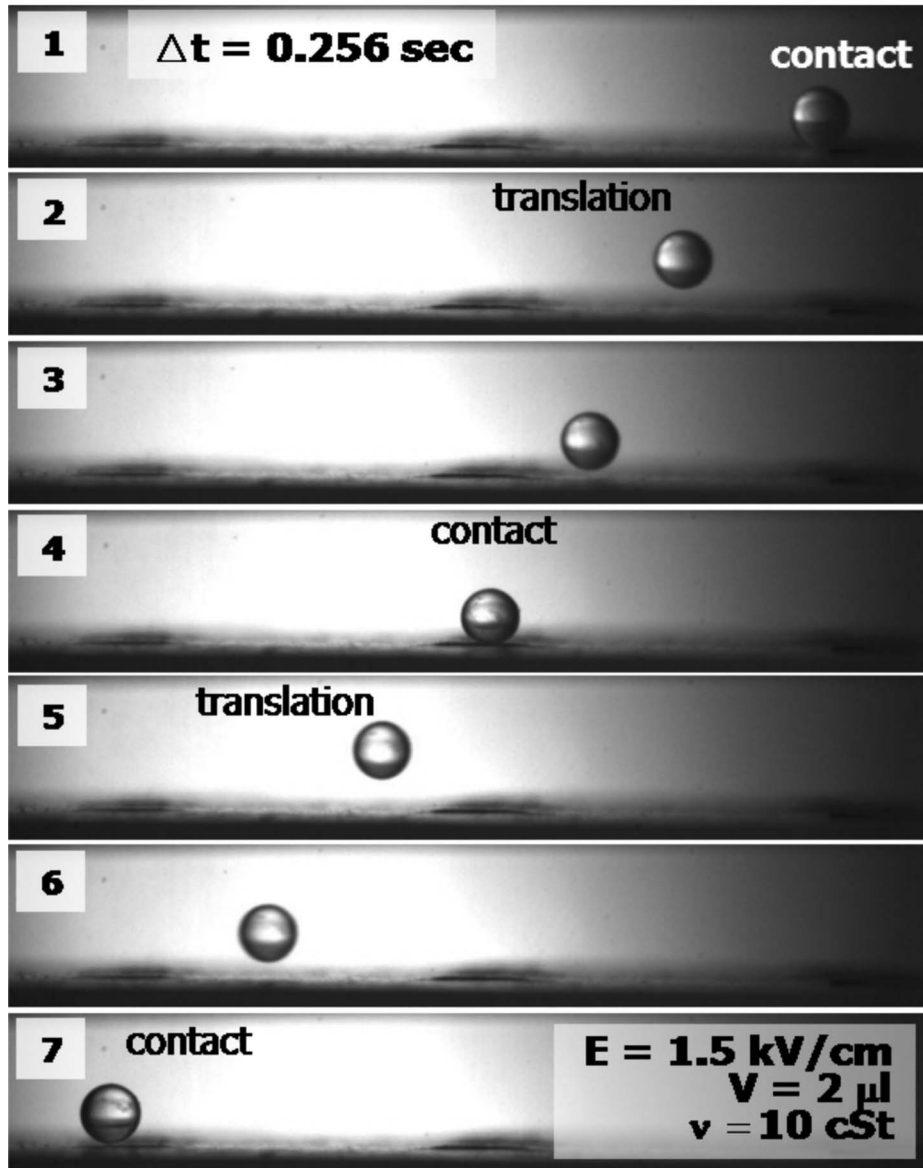
(b)

FIG. 7. The droplet is moved according to the electrical signal. (a) The droplet hops along the surface [URL: http://dx.doi.org/10.1063/1.3122299.1] (enhanced). (b) The sequential view of the behavior of a moving droplet (straight motion) (Δt=0.256 s, E=1.5 kV/cm, V=2 μl, and ν=10 cS).
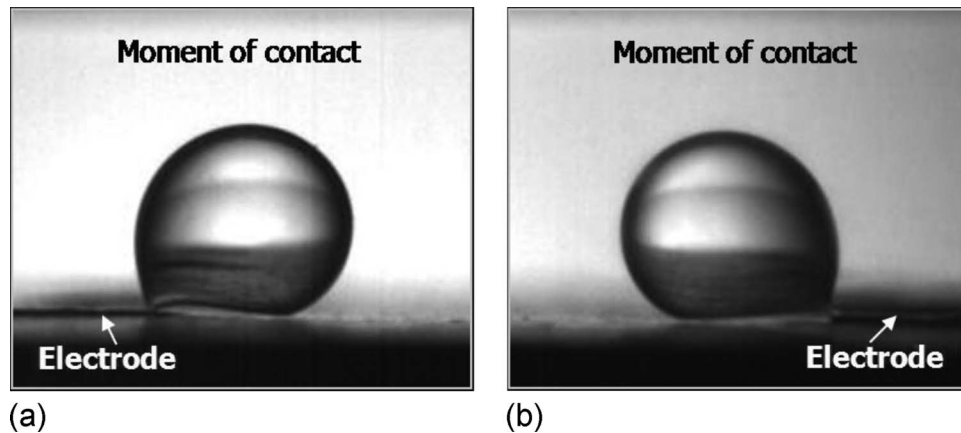
FIG. 8. The moment of contact of a droplet near the electrode [(a) negative electrode, (b) positive electrode]: The droplet is deformed into a cusped shape with a sharper tip.

B. Deformation of droplets near the electrode

The droplet experiences maximum elongations when the droplet is attracted toward the electrode and/or repelled from the electrode. The first elongation is the maximum elongation at the moment of contacting the electrode. As the droplet approaches the electrode, the droplet deforms into an elongated shape (Fig. 8). Because the droplet is a good conductor compared to the dielectric medium, the electric field becomes stronger in the gap between the electrode and the droplet surface. Thus charges are concentrated near the tip of the droplet facing the electrode, and the droplet is deformed into a shape with a sharper tip.[7] Therefore the maximum curvature of tip is formed at the moment of contact.

The second elongation occurs at the moment that droplet is repelled from the electrode (Fig. 9). At the moment of repulsion, the droplet is pushed outward very strongly. However, the tip of the droplet facing the electrode has oppositely polarized charges and it is pulled toward the electrode.

C. Electrocoalescence of two oppositely charged droplets

The electrocoalescence of two oppositely charged droplets was observed in detail using DI water with 50 vol % ink (Fig. 10). When two oppositely charged droplets approach each other due to Coulombic force, a liquid bridge is formed between them (Fig. 11). This liquid bridge plays a role as the electrical conduction path through which the electrical charges may be exchanged between the droplets. Immediately after the charge exchange, the droplet (a fluid body connected by the liquid bridge) becomes virtually neutral (if two droplets had the same amount of charges with different signs). However, it should also be noted that the fluid body of two droplets connected by a liquid bridge is subjected to electric polarization by the applied electric field. Thus, the electrical force is trying to pull the fluid body apart into two droplets again.

On the other hand, the surface tension force is trying to make the whole fluid body into a bigger spherical droplet. Therefore, if the electrical force is weaker than the surface tension force,
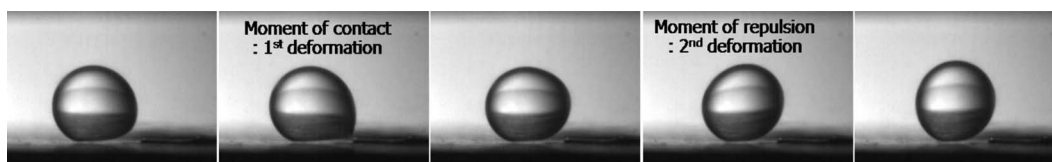

FIG. 9. The droplet experiences maximum elongations when the droplet is attracted toward the electrode and repelled from the electrode.
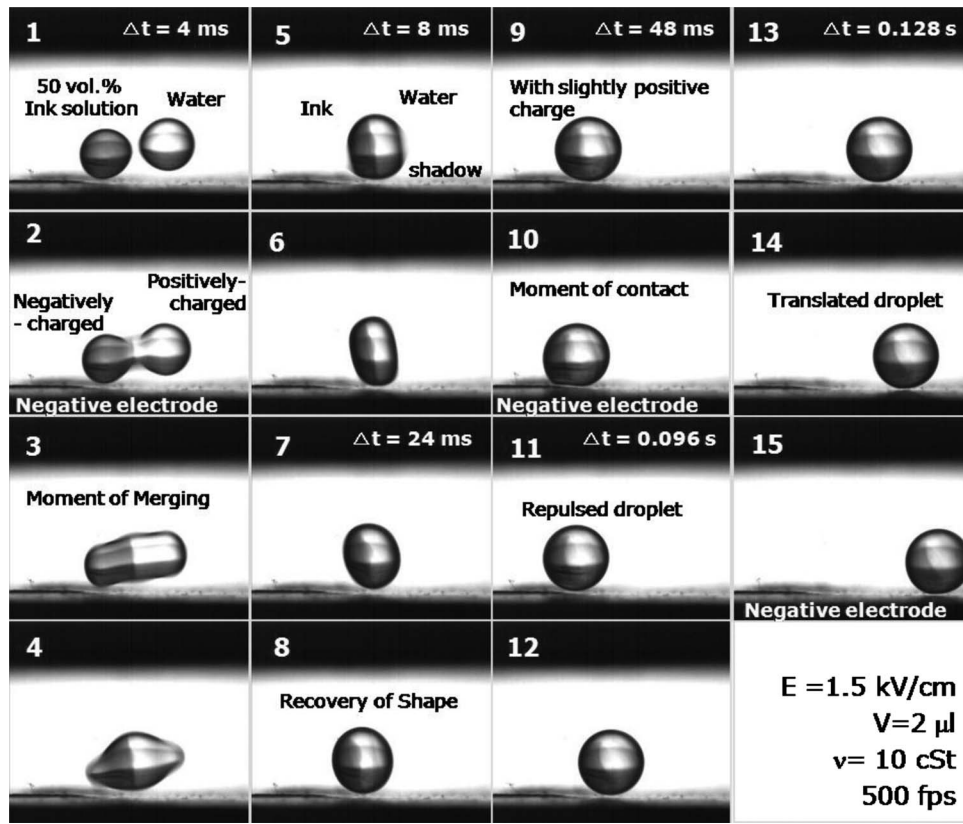
FIG. 10. [(1)–(9)] Side view of electrocoalescence and [(10)–(15)] repelled motion of the merged droplet. Gray colored: 50 vol % ink+DI water solution; white colored: Pure DI water droplet.

the droplets will be merged eventually into a bigger droplet. One example of this case is in Fig. 10. On the other hand, if the electrical force is stronger than the surface tension force, the fluid body will be broken into two droplets again. Then the two droplets are transported in the opposite directions, as shown in Fig. 11 (frames 4, 5, and 6).

Since the electrical force is approximately proportional to the square of the applied electric field strength, the most important parameter for determining whether the droplets are merged or not is the applied electric field strength at the moment of and after the liquid bridge formation. Merging is expected if the applied electric field after the liquid bridge formation is lower than a certain critical value. (The electric field does not have to be constant. We may have the same expectation if the electric field is lowered below the critical value at the moment.) In Fig. 10, the applied electric field was 1.5 kV/cm and the droplets were merged. On the other hand, in Fig. 11, the field was 4 kV/cm and the fluid body was broken into two droplets again.

The relative importance of the deforming electrical force to the restoring surface tension force may well be represented by the electrical Weber number, which is defined as $We = \varepsilon E^2 l / \gamma$, where l is the diameter of the undeformed merged droplet. When the applied electric field was 1.5 kV/cm, the Weber number was about 0.068 and the droplets were merged. However, when we increased the electric field to 3 kV/cm, two droplets started to be separated into two droplets. At this time, the Weber number was about 0.272. When we further increased the electric field to 4 kV/cm, the Weber number was about 0.484 and the merged droplet was broken into two droplets, as we have seen in Fig. 11. This tells us that there is a certain critical Weber number for breakup and that the electrical Weber number is the most important parameter for the merging dynamics of charged droplets under electric field.

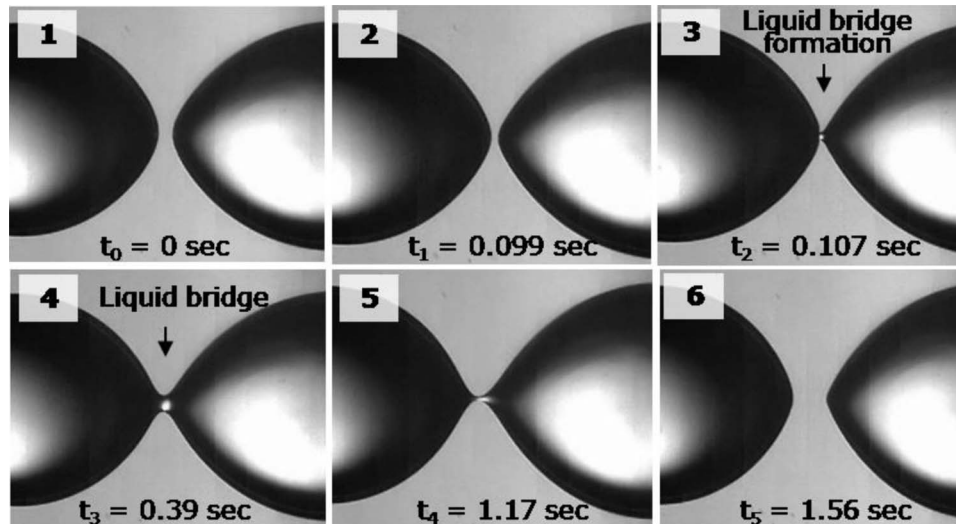When the merged droplet has zero net charge, it is not easy to transport the droplet to the

FIG. 11. The formation of a liquid bridge between two oppositely charged droplets in silicone oil medium (E =4.0 kV/cm, V=2 μl, and ν=50 cS).

desired position. So, it would be better to have nonzero net charge after merging. This can be easily achieved by using two droplets of different sizes. According to the previous work,[7] the acquired charge from the electrode is proportional to 1.59 power of the radius. We may exploit the scaling law. If the droplet is a perfect conductor with perfectly spherical shape, theoretical prediction of the exponent would be 2. For the spherical perfect conductors of the same size attached to the electrode, the surface charge density is proportional to the applied electric field. The conductor surface area is proportional to the square of radius, and the theoretical value 2 is explained. Deformability of the droplets and other effects may explain the difference between 1.59 and 2.

Even though the merging dynamics of charged droplets is explained qualitatively as shown above, its understanding is far from being sufficient for efficient control of droplets. Thus further study is definitely needed and experiments and numerical analyses will be performed as a continuing work.

D. Liquid bridge formation between two droplets

The close-up view of the liquid bridge at the moment of electrocoalescence was obtained (Fig. 11). As the droplets approach each other, the tips of droplets start to become sharp due to the concentrated charges (of opposite signs) near the tip (frame 2 of Fig. 11). A further decrease in the gap between two droplets results in a stronger electric field in the gap. This makes the tips sharper and more charges are concentrated near the tip. These concentrated charges cause stronger mutual attraction of the tips. Thus, when the initial gap is small enough, the sharp droplet tips are elongated to eventually meet each other and form the liquid bridge, as shown in frame 3 of Fig. 11. After the liquid bridge is formed, the charges are exchanged and the droplets can be merged or broken into two droplets again, depending on the applied electric field strength, as explained in Sec. IV C.

V. CONCLUSIONS AND ECOD NEXT STEP

We demonstrated the feasibility of a novel actuation method for manipulating conducting droplets by using ECOD. This method has several advantages over other methods. It causes less contamination of the chip substrate because a droplet is transported by hopping along the electric field line. Accordingly, surface treatment of the chip substrate is much simpler. Also, the droplet

transport is relatively fast: The average velocity is about 10 mm s^{-1}.[7] Therefore, we believe that ECOD-based microfluidics can be used as a tool for transporting a single microliter or nanoliter droplet without moving the medium fluid.

Further basic research and complete integration of the ECOD-based actuation method for droplet-based microfluidics should address many subjects, including individual manipulation, merging, sorting, breakup, and chemical reactions using droplets.

One potential use of droplet-based microfluidic devices is to encapsulate a varied population or library of molecules, cells, or particles into microreactors. This is possible because there is virtually no electric field inside a conducting droplet. When a conducting droplet is used as a tool for transporting bioparticles, such as DNA molecules, the droplet shields them from the strong external electric field. Technology for high-throughput droplet reactors continues to evolve and the pace of the development of new applications has increased.[1] Possibly, these various technologies can be realized by using ECOD.

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