Coalescence of Two Oppositely Charged Droplets at Constant Electric Potential

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Abstract – Electrocoalescence is an active technique in petroleum industry, formation of raindrop in cloud, and digital microfluidics. In the present work, electrocoalescence of two droplets under the constant electric potential in air was studied. Through this experiment, we found that the electrocoalescence process could be divided three phases; deformation, formation of liquid bridge, and merging. And the condition for formation of liquid bridge between two droplets was obtained. For the connection of experimental result in constant potential condition with general case in constant charge condition, relationship of charge and potential difference was deduced by numerical computation. In high electric potential case, flat interfaces after recoiling were observed. It was interpreted through a numerical simulation of electric field.

Key words: Electrocoalescence, Droplet, Microfluidics

1. Introduction

Electrocoalescence represents enhancement of coalescence of two droplets or enhancement of merging a droplet into interface of two immiscible fluids caused by DC or AC electric field [1]. Over the past decades, electrocoalescence has been studied as one of key mechanism of the formation of raindrop in cloud [2]. In the petroleum industry, electrocoalescence is also an important process for separation of water in crude oil [1]. Zhang *et al.* theoretically modeled a system of electrocoalescence considering hydrodynamic interaction between two droplets and introducing collision model [3,4]. According to their paper, the rate of collision significantly increases by imposing an external electric field and coalescence occurs more frequently for larger drops.

Recently, micro total analysis system (micro-TAS) taking the limelight, the importance of microfluidics, specially the importance of droplet based digital microfluidics, has increased [5-7]. Because of the possibility of chemical and biological applications of digital microfluidics, droplet actuation methods, such as electrowetting on dielectrics or dielectrophoresis, are developed [5-9]. And recently, a new method for droplet actuation has been developed, which is electrophoresis of charged droplets (ECD) [7,10-19]. In this method, droplets which are charged because of contact with electrode are controlled by the Coulomb force. Because ECD method can prevent

contamination of droplets and does not require any complex electrode structure, this method is expected to take advantage of an actuation method of droplets. And, in the ECD method, electrocoalescence has been renewal of interest as a way to merge the droplets. In microchannel, coalescence of droplets is also possible using variation of channel geometry without electric field or collision [20,21]. However, electrocoalescence has advantage that the droplets which we want could be merged actively at the time when we want. Therefore, recently, electrophoresis of charged droplet is suggested as a new actuation method of droplet for digital microfluidics system [7,10-19].

Miller *et al.* studied coalescence of oppositely charged falling droplets and investigated charge transfer between them experimentally [22]. And in 1968, G. I. Taylor modeled this problem as two circular soap films attached to ring shape electrode and theoretically investigated the shape of droplet using the force balance between capillary force and electric force [23]. After G. I. Taylor, there have been many experimental or theoretical studies. Priest *et al.* made an electrocoalescence system in the microchannel and showed that necessary voltage for coalescence is proportional to the distance between two droplets [24]. And Eow *et al.* studied the effect of angle between approaching direction and electric field and the effect of geometry of electrode [25].

When two oppositely charged droplets are contact, we expect they might merge because the one large drop has smaller surface energy then two separated droplets. However, coalescence of two contacting droplets does not always occur. Jung *et al.* reported that two oppositely charged droplets don't merge under high applied voltage (about 4 kV/cm) [11]. Independently from them, Ristenpart *et al.* also reported non-coalescence of oppositely charged droplets in high voltage case [26]. And they found that the non-coalescence phenomenon is caused by the direction of capillary force at the liquid bridge between two

droplets and they calculated the critical cone angle (which is 30.8°) [27,28].

The processes of electrocoalescence could be divided as **a)** approaching two droplets to each other, **b)** formation of liquid bridge between two droplets, and **c)** coalescence or recoiling [26,27,29,30]. In this work, first, we investigated the condition in which the liquid bridge is formed between two droplets under constant electric potential condition. Many researchers have studied this condition theoretically and experimentally [23,29,31]. In the theoretical point of view, the condition for the liquid bridge formation between two droplets is equivalent with non-existence condition of shape function of deformed droplets. In this work, to find the condition, the critical distance between two droplets and the critical voltage is measured just before formation of the liquid bridge. Second, flat interface after the recoiling is investigated. In high applied voltage case, after the recoiling occurred, flat interface is formed and it remains for milliseconds [26,27]. In this work, why this flat interface is formed and why it maintained metastably for a quit long time are interpreted through a numerical simulation.

2. Experimental Method and Material

In many case, electrocoalescence is done in another liquid phase such as silicon oil or crude oil. When two droplets are close under the electric field, the droplets are deformed to ellipsoidal shape. Facing ends of the droplets are more severely deformed, and eventually make Taylor's cone. So, near the tip, electric force and capillary force is dominant and hydrodynamic force could be negligible. Thus, the experiments could be conducted in air. The experimental setup appears in Fig. 1(a). To apply the electric field, high voltage power supply (TREK Model 610E) is used. The electric potential applied between two nozzles are 50~1000V. And high speed camera (Photron[®] FASTCAM 1024 PCI) is used for observation of the coalescence process. To make droplets, syringes and stainless steel syringe nozzles are used. To diversify the radius of water droplets, two different sizes of nozzles were used, one is 20 gauge of which outer diameter is 0.90 mm and another is 26 gauge of which outer diameter is 0.46 mm. Basically, the droplets are made with deionized water of which surface tension is 0.072 N/m. Additionally, to find out the effect of surface tension, cetylpyridinium chloride (CPC), which is a non-ionic surfactants, was used. The surface tension of the liquid is measured using tensiometer (LAUDA Automatic Tensiometer TD2).

In this study, experiments are carried out in two ways. One is named electrocoalescence of non-deformed droplets (ECNDD). In this experiment, the electric voltage is suddenly applied to merge non-deformed (partially spherical shaped) droplets. In digital microfluidics system, droplet coalescence on demand is necessary. Thus, electric field is applied at the specific time, and on the specific place [24]. Therefore, electrocoalescence of non-deformed droplet is studied using suddenly applied electric field. Throughout this experiment,
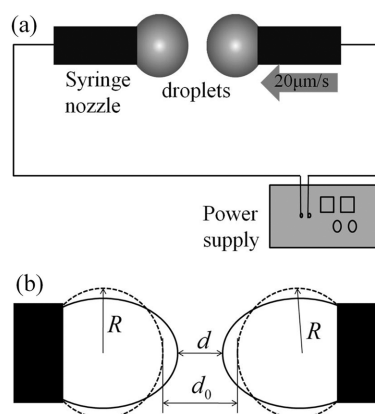
Fig. 1. Experimental setup and definition of geometry factors (a) Two equal sized droplets are formed at tips of stainless steel nozzle. In the "electrocoalescence on non-deformed droplets", step functional electric field is applied at stationary spherical droplets and in the "electrocoalescence of deformed droplets", one nozzle is approached to another with constant velocity applied constant electric potential. (b) Dashed line represents shape of non-deformed spherical droplets and solid line represents deformed droplets. R is radius of spherical droplets, d_0 is initial distance of two droplets, d is distance between two deformed droplets.

initial distance between two droplets could be measured. But it is difficult to measure the critical distance through this experiment. Because the droplets deform rapidly, inertial force influence on the deformation of droplets. Therefore, in this case, it is impossible to measure the critical distance which is a boundary of the equilibrium shape and non-equilibrium shape. Another experiment was required to measure critical distance.

Second experiment is electrocoalescence of deformed droplets (ECDD). In this experiment, as shown in Fig. 1, a droplet is approached very slowly to another droplet after applying DC electric field. So the liquid bridge is formed between two droplets which are already deformed to ellipsoidal shape under the applied electric field. In many cases such like rain drop formation or purification process of clued oil, droplets are merging after deformed under the influence of electric field. In this experiment, a motorized stage (OptoSigma[®] SGSP20-20) is used to make constant approaching velocity (20 μm/s). Through this experiment, the critical distance between two droplets could be measured.

3. Result and Discussion

3-1. Process of electrocoalescence and critical distance

Fig. 2 and 3 are high speed image of electrocoalescence. As you can observe in Fig. 2, if electric potential is not high enough or initial distance between two droplets are not close enough, droplets are not merging and they are oscillating because of inertial force (column (1) of Fig. 2). And if electric potential between two droplets are high enough, droplets are recoiling after touching and flat interface is formed (column (3) of Fig. 2). This flat interface will be discussed in

(1) 600V (2) 800V (3) 900V

(a) Deformation

t=0 t=0 t=0

t=0.83ms t=0.83ms t=2.5ms

(b) Formation of liquid bridge

t=1.67ms t=1.67ms t=3.33ms

(c) Vibration (d) Merging (e) Flat interface

t=2.50ms t=1.83ms t=3.50ms

t=4.17ms t=2.0ms t=3.67ms

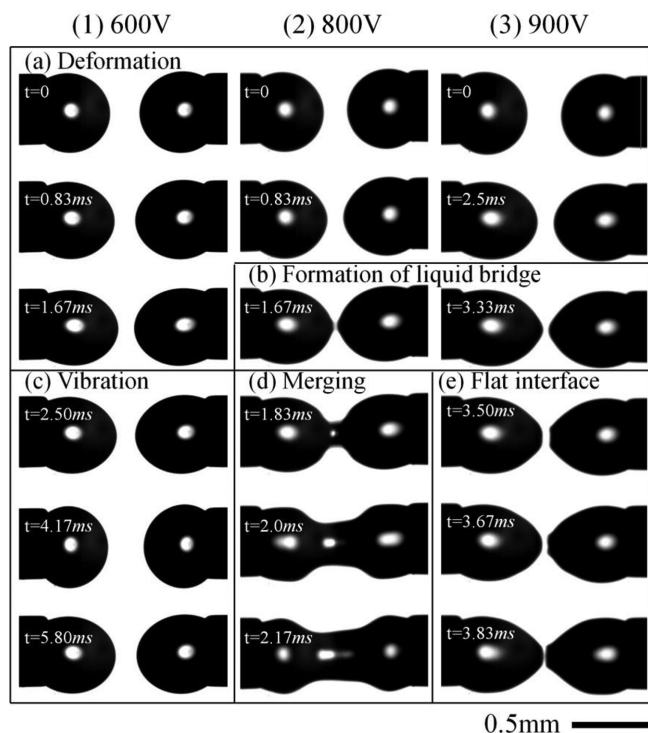t=5.80ms t=2.17ms t=3.83ms

0.5mm

Fig. 2. Deformation and coalescence of non-deformed droplets. Frame rate is 6,000 frames per second. Column (1) Applied voltage is 600V but distance is not small enough. So the droplets are not merging. Column (2) Applied voltage is 800V and distance is smaller than the critical distance. So the droplets are merging. Column (3) Applied voltage is 900V and the cone angles are sharp. So the droplets are recoiled and flat interface is formed.
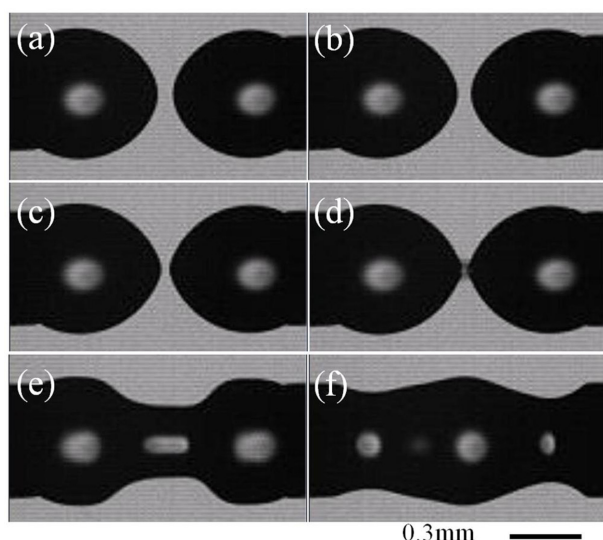

(a) (b)

(c) (d)

(e) (f)

0.3mm

Fig. 3. High speed images of ECDD(electrocoalescence of deformed droplets). Initial radius of droplets is 0.3 mm, applied voltage is 650V and frame rate is 27,000 frames per second. (a) t=-0.37 ms, (b) t=-0.185 ms, (c) t=-0.037 ms, (d) t=0, (e) t= 0.185 ms and (f) t=0.37 ms. (a,b) are deformation phase, (c,d) are liquid bridge formation phase, and (e,f) are merging phase.

section 3.3. Except these two case, electrocoalescence could be divided three phases, which are deformation, formation of liquid bridge, and merging (column (2) of Fig. 2 and Fig. 3).

3-1-1. Phase 1. Deformation

At the beginning, each droplet is deformed relatively slowly and the droplet changes its overall shape from a sphere to an ellipsoid (Fig. 2(a) and Fig. 3(a, b)). In this phase, capillary force and electric force achieve equilibrium and the deformed shape is stable. Therefore, if the applied potential is not enough to merge in ECNDD case or if the approaching is stopped in ECDD case, the deformation of the droplet is stopped and the deformed ellipsoidal shapes are maintained.

3-1-2. Phase 2. Formation of Liquid Bridge

If the applied voltage is high enough in ECNDD case or droplets are approaching in ECDD case, after the deformation phase, deformations are going very fast at the tip of droplets and a liquid bridge is formed very quickly between two tips of droplets (Fig. 2(b) and Fig. 3(c, d)). In this phase, the droplets do not make change of overall shape, but make liquid bridge just on tips of each droplet. Because the electric force is overwhelming the capillary force at the tips of droplets, the shape of the tip is not stable, and the liquid bridge is formed regardless of the approach of droplets in ECDD case.

3-1-3. Phase 3. Merging

After the liquid bridge is formed between two droplets, charge on that region is neutralized. The droplets are merged spontaneously caused by inertial force and capillary force. In this phase, the liquid bridge grows thick and two droplets are merging eventually (Fig. 2(d) and Fig. 3(e, f)).

This phase division for electrocoalescence is consistent with previous studies [29,30,32]. In the theoretical point of view, a classification criterion of phase 1 and phase 2 is the existence of a static solution of the differential equation for the deformed shape of droplets [23,29,30]. If the distance between two droplets (d) is smaller than the critical distance (d_{crt}) or the potential difference between droplets (V) is larger than critical voltage (V_{crt}), then there are no static solution of the shape equation and it means that the droplets cannot exist separately. G. I. Taylor simplified this problem to a linearized force balance equation for the deformed shape [23]. He assumed that i) distance between droplets are very close ($d_0 \ll R$), ii) the interface is equipotential surface, iii) deformation occurs only in some parts of the facing. From these assumptions, Taylor obtained asymptotic solutions of critical distance and critical voltage.

$$d_{crt} \approx 0.50d_0, \quad V_{crt} \approx 0.38d_0\sqrt{\frac{\gamma}{\varepsilon_{out}R}}. \quad (1)$$

In the experimental point of view, another important criterion to differentiate phase 1 and phase 2 is the stability of droplet shape. In the deformation phase, shapes of two droplets are stable but in the liquid bridge formation phase the shapes of two droplets are not stable. So the critical distance means the minimum distance between two droplets without touching each other under the applied electric field. In ECNDD case, the droplets deform rapidly because of sudden electric field and inertial force affect to the coalescence process.

Therefore, droplets do not reach equilibrium shape and critical distance could not be measured in ECNDD method. So, to measure critical distance, we need ECDD method in which a droplet approach to another very slowly and the droplets reach equilibrium shape before phase 2. Thus, if the approach of droplet is stopped, deformation is also stopped in phase 1. But, in phase 2, the liquid bridge is formed very quickly, even though the approach of droplet is stopped.

An experimental method to divide first phase and second phase is to measure the deformation velocity. In the deformation phase, deformation velocity is mild, about several decade millimeters per second. But, in the liquid bridge formation phase, the deformation velocity is very fast, more than hundreds of millimeters per second (Fig. 4). Thus, through measuring the distance between tips of two droplets (d), critical distance (d_{crt}) which divides phase 1 and phase 2 could be determined. Discontinuity in the graph of Fig. 4. is caused by discontinuous motion of stepper motor in the motorized stage to make constant velocity.

Dielectric permittivity of water is much bigger than that of surrounding air. Therefore, applied electric potential between two electrodes is almost same with that between two tips of the droplets. So V/d is more relevant than $V/2R$ as the characteristic field strength, where V is applied voltage. Thus, electrical Weber number should be defined as

$$We \equiv \frac{\varepsilon_{out}(V/d)^2}{(\gamma/R)} = \frac{\varepsilon_{out}V^2 R}{\gamma d^2}, \quad (2)$$

where ε_{out} is dielectric permittivity of air, and γ is surface tension of the droplets.

Fig. 5 represents experimental result about $\varepsilon_{out}V^2$ vs. $\gamma d^2/R$. The critical distance (d_{crt}) in this results is measured from the ECDD method and the initial distance (d_0) in the inset figure is measured from the ECNDD method. As you can see in this figure, $\varepsilon_{out}V^2$ vs. $\gamma d^2/R$ has a linear relationship regardless of droplet radius and surface tension, and their slops are 3.5 for critical distance in ECDD and 0.31 for initial distance in ECNDD. It means that
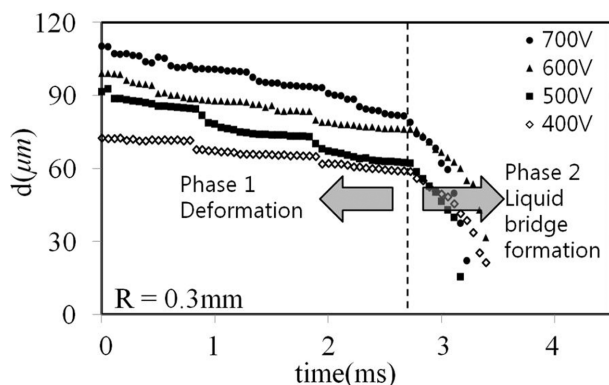

Fig. 4. The distance between droplets (d) shrinks dramatically after critical point. That point divides phase 1 and phase 2 and, at that time, the distance could be defined as critical distance (d_{crt}). Discontinuous of graph is caused by movement of stepping motor.
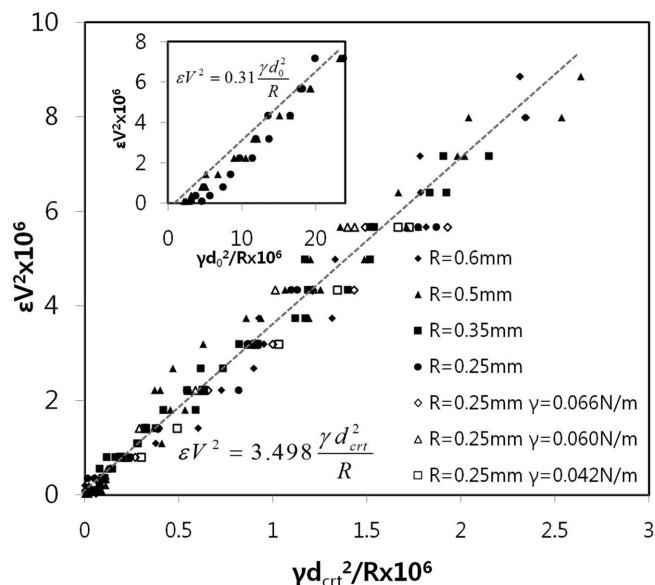
Fig. 5. The experimental data are plotted as the form of εV^2 vs. $\sigma d_{crt}^2/R$, and εV^2 vs. $\sigma d_0^2/R$ (inset). Two variables have linear relationship and their slops are 3.5 and 0.31, respectively. The slops represent We_{crt} and We_0.

$$We_{crt} = \frac{\varepsilon_{out}V_{crt}^2 R}{\gamma d_{crt}^2} \approx 3.5 \text{ and } We_0 = \frac{\varepsilon_{out}V_{crt}^2 R}{\gamma d_0^2} \approx 0.31. \quad (3)$$

As well known, the electrical Weber number is a ratio of capillary force to maintain its shape and electric force to deform it. So we can say that when two oppositely charged droplets are approaching under applied electric field, the liquid bridge is formed between them if the distance (d) is just about $V\sqrt{\varepsilon_{out}R/3.5\gamma}$, and $d_{crt} \approx 0.30d_0$. And if we want to merge two stationary droplets, required voltage (V_{crt}) is more than $d_0\sqrt{0.31\gamma/\varepsilon_{out}R} = 0.56d_0\sqrt{\gamma/\varepsilon_{out}R}$.

Comparing with eq. (1), which is derived theoretically by G. I. Taylor, the critical voltage is larger than that and the critical distance is smaller than that. This difference between theoretically expected value and experimentally measured value is caused by perfect conductor assumption and small distance assumption ($d_0 \ll R$). Although dielectric constant of deionized water is much larger than that of air, conductivity of water is very low comparing with general conductor. Thus, potential difference between tips of two droplets is a little lower than applied voltage on the electrode. Another reason is the small distance assumption that Taylor used. In this experiment, distance between two droplets is quite large so the small distance assumption doesn't hold. Because of these two reason, the experimental values of d_{crt} and V_{crt} are different with the theoretical values.

3-2. Charge of droplets and potential difference

In many cases, electrocoalescence occurs in condition that the charge of droplets is conserved. The formation of rain droplet, electrocoalescence process in purification of crude oil and droplet merging in ECD are in the charge conserved condition. However, in this study droplets attached to the electrodes and the electric potential of the droplets are maintained constantly. Constant voltage condition

can be applied electrocoalescence in microchannel but it is a relatively rare condition comparing with constant charge condition. Although a freely moving droplet has the constant charge, droplet could be considered as a equipotential conducting material because the charge relaxation time of pure water is very short ($\tau_c = \varepsilon_{water}/\sigma_{water} \approx 1.2 \times 10^{-4} s$). Therefore, droplet which has constant charge and has some position could be considered as an equivalent droplet which has a constant electric potential.

Fortunately, analytic solution is available for the case of two charged conducting spheres in a uniform field [33]. M.H. Davis was solved this boundary value problem using bispherical harmonics. Rather complicated, the electric potential of two equal size spherical conductor which are oppositely charged could be written like that

$$V_1 = -V_2 = \frac{V}{2} = P_{11}Q_1 + P_{12}Q_2 \ . \tag{4}$$

Hear V_1 and V_2 are potential of each conductor and potential difference between two conductor is $V = 2V_1 = -2V_2$. The charge of each conductors are Q_1 and Q_2, and $Q_1 = -Q_2 = Q$ in this case. The coefficients of induction, P_{ij}, could be written

$$P_{11} = \frac{C_{11}}{C_{11}^2 - C_{12}^2}, \quad P_{12} = \frac{-C_{12}}{C_{11}^2 - C_{12}^2} \ . \tag{5}$$

And the capacitance, C_{ij}, are

$$C_{11} = 2\varepsilon_{out} a \sum_{n=0}^{\infty} \frac{e^{(2n+1)\mu_1}}{e^{2(2n+1)\mu_1} - 1}, \quad C_{12} = -2\varepsilon_{out} a \sum_{n=0}^{\infty} \frac{1}{e^{2(2n+1)\mu_1} - 1}, \tag{6}$$

where a bispherical scale factor $a = \sqrt{(R + d_0/2)^2 - R^2}$, and a bispherical coordinate of spherical conductor surface $\mu_1 = \ln\left(\frac{R + d_0/2 + a}{R}\right)$.

The analytic solution derived by Davis is a general solution covering different size, various charge and external electric field. However, there is a little discomfort for practical use because it contains infinite series. So we conducted a numerical computation for this problem. Domain of computation and boundary conditions are represented in Fig. 6. The droplets are considered as perfect conductors so the electric potential of spheres surfaces are $+0.5V$ and $-0.5V$, respectively. To minimize wall effect, the computational domain is made large enough, $D = 5\sim10R$ and $L = 10\sim20R$. And the radius of spherical conductor (R) and distance between two conductors (d) are 0.25~1 mm and 0.05~0.4 mm, respectively. Potential difference between two conductor is 100~1000V. All computations were performed by using Comsol Multiphysics version 3.5.

From the electric potential distribution obtained by numerical computation, charge amount of a conductor could be calculated through integration of surface charge density.

$$Q_1 = \int_{S_1} \sigma_e dS = \int_{S_1} \varepsilon_{out} \mathbf{n} \cdot \nabla \phi dS \ , \tag{7}$$

where S_1 is surface of conductor which has positive potential, σ_e
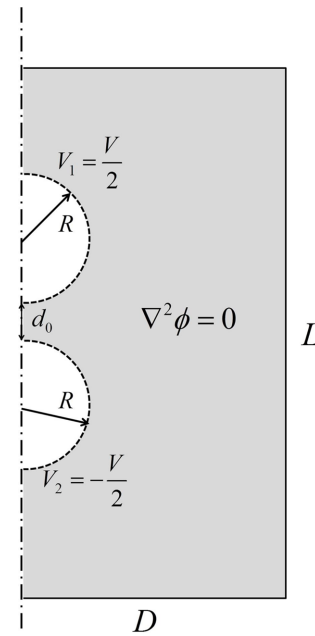

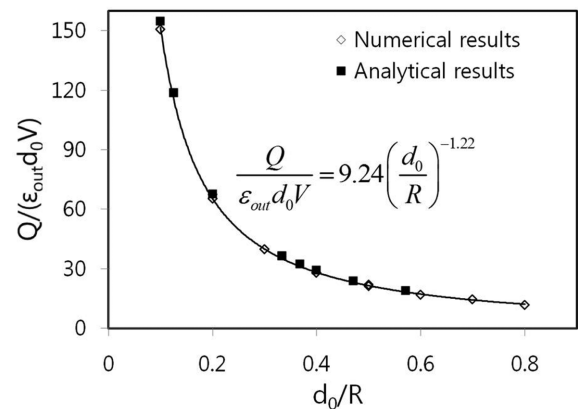Fig. 6. Domain of computation and boundary conditions.

Fig. 7. Relation of charge and potential difference. All results are non-dimensionalized and numerical results are consistent with the analytical results.

is surface charge density and $\mathbf{n}$ is outward normal vector of surface of the sphere. Results of computation could be non-dimensionalized using two dimensionless number, d_0/R and $Q/(\varepsilon_{out}d_0V)$. As you can see in Fig. 7, the charge amount of droplet has linear relationship with the potential difference between two droplets. And the relation could be written like that

$$Q = 9.24\varepsilon_{out}Vd_0\left(\frac{d_0}{R}\right)^{-1.22}, \tag{8}$$

and this is consistent with the analytic result derived by Davis.

3-3. Flat interface after recoiling

As shown in Fig. 8, droplets are not merging and make flat interface between them, in high voltage (above 800V) case. This flat interface is formed after touching and recoiling of two tips of the
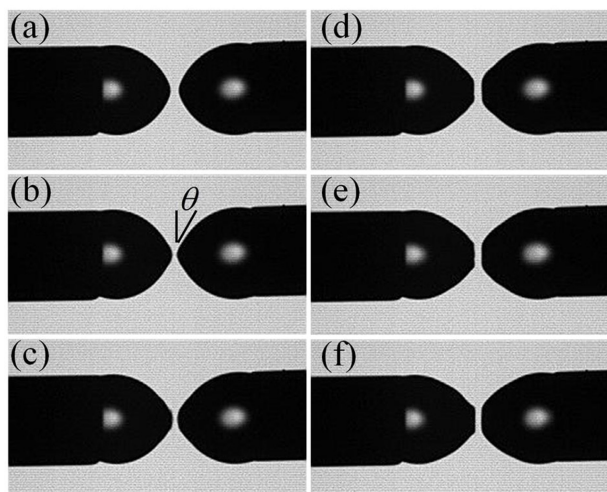
Fig. 8. Flat interface after recoiling. Applied voltage is 900V, drop radius is 0.5 mm and the frame rate is 27,000 frames per seconds. Time step between each picture is 74.08 μs.
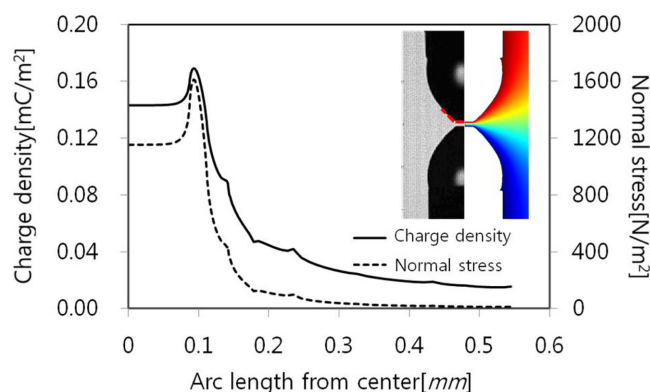

Fig. 9. Solid line and dotted line represent charge density and normal component of Maxwell stress along the red dotted line in inset figure. Left side of the inset figure is high speed image 278 μs after the liquid bridge is broken and right side represent electric field from numerical simulation. Applied voltage is 900V and initial drop radius is 0.5 mm.

droplets. Both in oil and in air, if the applied voltage is high enough, non-coalescence of oppositely charged water droplets was observed through experiments [11,26,27]. However, if droplets are freely moving in outer media instead fixed at the electrode, recoiled droplets go away from each other. Therefore, the flat interface after recoiling could be observed only in case which the droplets are fixed on electrode [27].

After touching of two Taylor cone, all charge near the cone is neutralized in an instant and the capillary force is the only dominant force. Thus, according the direction of capillary force, the droplets can merge or fail to merge. And according to J. C. Bird *et al.*, the direction of capillary force is determined by cone angle, θ [27,28]. In other words, if the cone angle is less then critical angle when the applied voltage is mild, two droplets can merge and in the other case when the applied voltage is high enough, two droplets fail to merge.

After two tips of droplets are touching and recoiling, the flat interface is formed momentarily (Fig. 8(c, d)). The charge relaxation time ($\tau_e = \varepsilon_{in}\varepsilon_0/\sigma_e$) of deionized water is about 100 μs. So during that time, this region is electrically neutral. But even though after that time the flat interface is maintained over hundreds of microseconds (Fig. 8(e, f)). The viscous time scale ($\tau_h = \rho_{in}d^2/\mu_{in}$) is much larger than the charge relaxation time but capillary time scale ($\tau_s = \sqrt{\rho_{in}d/\gamma}$) has same order with the charge relaxation time. Thus, we can say that this flat interface after recoiling is caused by equilibrium of electric force and capillary force. To verify that, numerical analysis was performed using experimentally obtained shape.

Fig. 9 shows surface charge density and normal component of Maxwell stress ($\mathbf{F}_{e,n}$) along surface line from the center.

$$\mathbf{F}_{e,n} = \mathbf{n} \cdot \mathbf{T}_e \cdot \mathbf{n}, \tag{9}$$

where $\mathbf{T}_e$ is Maxwell stress tensor, $\mathbf{n}$ is outward normal vector of surface. As you can see this figure, on the flat interface the electrostatic force is not large enough to pull the liquid behind the

interface. However, at the edge of the flat interface density of surface charge and normal stress have maximum value. Thus, the electric force is maximum at the edge, and the direction of force at the edge has both vertical and horizontal component which is pointing in the direction of increasing radius of flat interface. Because of the vertical component of the electric force, two droplets are merged eventually. And because of the horizontal component of the force at the edge, flat interface is maintained and even increased until merging.

4. Conclusion

In present work, we investigated the electrocoalescence of two equal sized droplets which are at constant electric potential. The electrocoalescence process of two droplets could be divided three phases; deformation, formation of liquid bridge, and merging. It is confirmed experimentally that applied voltage required to make liquid bridge between two droplets is proportional to distance of the droplets in both deformed droplets and non-deformed droplets case. And this is consistent with previous works [1,24,27].

When the liquid bridge was formed between droplets, the critical Weber number (We_{crt}) was about 3.5. In practical point of view, it seems like there is no meaning because it is impossible to measure electric potential between two droplets in bulk system such as formation of raindrop or purification process of crude oil. But this number is a criterion for the formation of liquid bridge. From a theoretical perspective, therefore, this number indicates a condition for the formation of liquid bridge on the interface of droplet which is already deformed by electric field. The initial Weber number (We_0) gives us more practical information than the critical Weber number. If we know the size of droplets and distance between them, this number indicates how strong electric field is required to make electrocoalescence in the given water in oil system, not only a bulk system but also microfluidic system.

For the connection of constant potential condition of this work to the constant charge condition of general case, the relationship between the charge and potential difference was deduced. Although there is analytical solution for the more general problem, for ease of use, numerical results were derived. Lastly, we analyzed the flat interfaces after recoiling of droplets. The flat interfaces are metastable because the capillary force and electrostatic force achieve equilibrium on the interface and at the edge. We interpreted it by a numerical simulation for the electric field on the flat interface after recoiling.

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