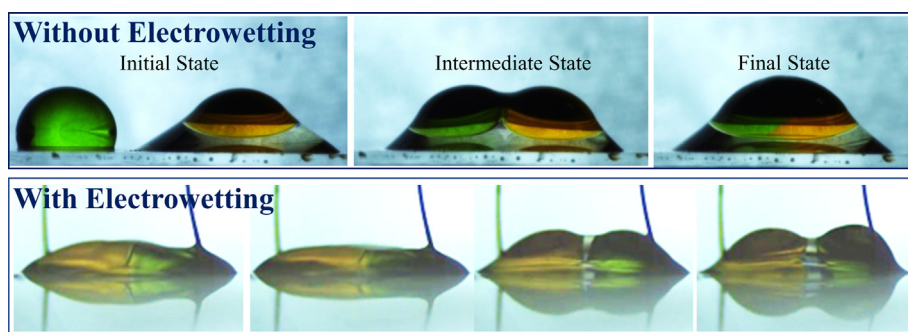Regular Article

# Effect of electrowetting induced capillary oscillations on coalescence of compound droplets

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GRAPHICAL ABSTRACT



ARTICLE INFO

ABSTRACT

*Hypothesis:* Coalescence time depends on the drainage rate of the fluid-bridge separating the droplets. Drainage rate is determined by external forcing and properties of the surrounding fluid. Modulating external forcing using electrowetting induced interface motion should allow control of the drainage rate, thereby affecting the coalescence time. Hence, quick coalescence or prolonged non-coalescence can be obtained for compound droplets on the microfluidic lab-on-chip systems.

*Experiments:* Using high-speed imaging, we have investigated the effect of electrowetting induced capillary oscillations on the coalescence of compound droplets consisting of water core encapsulated in an oil shell. A systematic study was performed by varying the shell viscosity and actuation parameters (i.e. amplitude, frequency and waveform).

*Findings:* For actuated interface, we observed specific regimes of coalescence or non-coalescence, whereas in absence of actuation, coalescence was observed in finite time. Non-coalescence was attributed to the continuous modulation of the oil-bridge width, which was caused by the interface motion. Oil-bridge width modulation was seen to be dependent on the amplitude and shape of the excited capillary modes (axisymmetric and non-axisymmetric). These modes were tuned by the actuation parameters. This is the first report of controlling coalescence dynamics by using electrowetting induced interface motion.

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## 1. Introduction

Compound droplets are formed by enclosing the sample droplet in an immiscible oil-shell [1–3]. This encapsulation allows us to manipulate samples and carry out reactions in an oil phase thereby preventing biofouling, evaporation, sample contamination and hysteresis. Several authors have explored manipulation of compound droplets through electrowetting [4,5]. Electrowetting is a commonly used technique to actuate liquids on microfluidic platforms. In a compound droplet based microfluidic platform, merging of two droplets (cores) co-encapsulated in a single oil-shell is

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slowed down due to the difficulty in draining the thin layer of oil between the two cores. Coalescence enhancement for drops on solid substrate or in micro-channels has been attained by applying electric field [6,7], by controlling flow pattern in sheared suspensions [8], by changing size of drops [9] or by using patterned electrodes [10]. Electro-coalescence uses electrostatic attraction for achieving faster coalescence [11,12] by applying opposite charges to two coalescing droplets through biasing electrodes [13,14]. Study of coalescence dynamics [15] is also important for applications like de-emulsification [16], mixing [17] and chemical reactions [18]. Effect of coalescence and interface dynamics on mixing enhancement has been studied using electrowetting on normal [19,20] and on super-hydrophobic surfaces [20].

Most coalescence enhancement techniques require separate electrodes or special structures and hence are difficult to integrate with existing electrowetting based device designs. Effect of electrowetting actuation has been investigated for coalescence of compound drops on liquid infused surfaces [21]. In that study, the drainage rate decreased and the coalescence time increased with increase in voltage for high frequency (10 kHz) electrowetting. This was attributed to the reduction in contact angle of aqueous drops by electrowetting. Contact angle reduction caused the interaction point of the aqueous drops to fall close to the oil meniscus. Thus, a thin layer of oil was pulled between the aqueous droplets which caused increase in the coalescence time. The previous study was however limited to an actuation regime (i.e. high frequency) where interface oscillations are completely suppressed.

Low frequency AC actuation which leads to large interface oscillations, have been studied in detail [25–27]. Understanding interface dynamics is important for several applications such as switches [28], displays [29] and lenses [30,31]. In lab-on-chip devices, interface oscillations have been used to enhance mixing [32–36]. In this work, we study the effect of electrowetting induced low frequency interface oscillations on the coalescence of compound droplets. We report regimes of actuation parameters (i.e. voltage and frequency), where coalescence in finite time or extended non-coalescence was observed. We elucidate how modulation in entrapped oil-bridge width leads to observance of non-coalescence. We explain how oscillation mode shape, shell viscosity, and contact line hysteresis regulate the oil-bridge width modulation and hence govern these regimes. Extended non-coalescence is attributed to the electrowetting induced capillary oscillations which repeatedly pushes the two cores away.

Karpitschka et al. [22–24] have studied the coalescence dynamics of miscible but different and non-identical liquids. They proposed an entirely different mechanism based on difference in surface tension and initial three phase contact angle of two miscible liquids. Marangoni flow developed during the coalescence delayed its completion. But for droplets of similar liquids, they suggested that instantaneous coalescence should occur. However, by inducing capillary oscillations using electrowetting, we were able to achieve non-coalescence between two core-droplets of the same liquid. In literature, similar non-coalescence has been observed for drops bouncing on vibrating liquid bath [37] due to entrapped air film and for Leidenfrost drops [38] due to the developed vapour film.

## 2. Materials and methods

### 2.1. Device fabrication and experimental setup

Devices were fabricated on Borofloat glass wafers. Transparent actuation electrodes were fabricated using ITO (indium tin oxide). A dielectric layer of SU8-2005 was deposited by spin coating at 3000 rpm for 30 s to get a thickness of $5.0 \pm 0.3$ $\mu$m. The thickness was measured using Dektak-XT surface profiler. Teflon® (AF-2400) solution (DuPont) was spin coated at 4000 rpm for 40 s and hard baked at 180 °C to form the topmost hydrophobic layer. The thickness of the Teflon layer was measured to be $170 \pm 30$ nm. A thin steel wire having 100 $\mu$m diameter was used to ground the core droplets from top.

A National Instruments multifunctional card (NI-DAQ 6363) was programmed to provide sinusoidal actuation voltage. The DAQ card simultaneously provided a trigger voltage (5V pulse signal) for synchronizing the video capture. The high-speed camera (Photron SA4 Fastcam) was operated at frame rates varying from 250 to 2000 fps. A high voltage amplifier (Trek 2205) with gain 50 was used to amplify the actuation voltage.

### 2.2. Experimental parameters

The configuration of the static compound drops depends solely on the relative interfacial energies and the disjoining pressures. For the present case, Neumann law and triangle inequality [1,5] are not satisfied and hence, the air-water-oil contact line is not formed. The spreading coefficient (S) is given by, $S = \gamma_{aw} - \gamma_{ow} - \gamma_{ao}$ where $\gamma_{ow}, \gamma_{ao}$ and $\gamma_{aw}$ are the surface tension of oil-water (35 mN/m [39]), air-oil (19.7 mN/m [40]) and air-water (72 mN/m) interfaces. The spreading coefficient is positive, implying a thin film of oil will cloak the water droplet. Surface energy calculations imply that a thin film of silicone oil will be present between the water droplet and Teflon coated substrate. Using Young's equation, the surface tension of Teflon-oil interface ($\gamma_{os} = 4.3$mN/m) and Teflon-water interface ($\gamma_{ws} = 48.5$mN/m) were obtained by using measured contact angle of water on Teflon as $113° \pm 3°$ and oil on Teflon as $37° \pm 3°$ [41,5]. The properties of silicone oil used for forming the encapsulation shell are tabulated in Table 1. Other parameters related to the compound droplet configuration used in these experiments are given in Table 2.

## 3. Experimental results

### 3.1. Coalescence in absence of actuation

The side-view images captured for coalescence of non-actuated compound droplets are shown in Fig. 1. Two water drops each of 5 μl volume were used as cores in this study. A 5 μl shell of 5 cSt silicone oil (Sigma Aldrich, see Table 1 for properties) was added on the first water droplet such that the oil-shell boundary touched the second water droplet. Upon contact, the oil-shell spreads to

**Table 1**
Properties of silicone oil used in the experiments.

| Viscosity ($\mu_o$) | Refractive index (n) | Surface tension ($\gamma_{ao}$) | Density ($\rho_o$) | Dielectric constant ($\epsilon_o$) |
|---|---|---|---|---|
| $5 \times 10^{-6}$ m²/s | 1.397 | 19.7 mN/m | 0.918 kg/m³ | 2.59 |

**Table 2**
Measurements of different parameters used in experiments.

| | |
|---|---|
| Radius of water core drop, $R_w$ | $1.27 \pm 0.04$mm |
| Radius of oil shell drop, $R_o$ | $2.5 \pm 0.04$mm |
| Contact angle of water drop on substrate | $113° \pm 3°$ |
| Contact angle of compound drop on substrate | $47° \pm 3°$ |
| Ridge height of oil from side view, $h_r$ | $0.8 \pm 0.04$mm |
| Hysteresis of oil-air interface | $10–15°$ |
| Compound drop height, $h$ | 1.8mm |