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# Critical AC frequency for stable operation of electrowetting-driven optofluidic devices with polymeric electrolyte solutions [†]

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## Abstract

EWOD (electrowetting on dielectric) is a viable scheme to drive optofluidic devices that utilize the liquid interface as a refractive surface. Although AC (alternating current) voltage is often applied to an electrode in EWOD devices to improve electrical characteristics, it may cause oscillations of liquid interfaces which can be detrimental to the system's performance as an optical device. Here we experimentally identify the origin of the interfacial oscillations of polymeric electrolyte solutions by observing the dynamic responses of contact lines as a function of the AC frequency. Penetration of small charged matters into the dielectric film of the opposite polarity is shown to induce the voltage relaxation that leads to deterioration of electrowetting performance. Measuring the relaxation time scale which is different for each polarity allows us to find the critical AC frequency that ensures stable interface control.

*Keywords*: Electrowetting-on-dielectric; Frequency; Interfacial oscillation; Polymeric electrolyte

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## 1. Introduction

When the electrical potential difference is established between a solid electrode and a sessile liquid drop, the contact angle is reduced from the value without the potential difference, a phenomenon originated from the electrowetting. The classical Young-Lippman equation relates the applied voltage to the contact angle change [1]. It is often used in the form of electrowetting on dielectric (EWOD), where a thin dielectric layer covers the electrode. With the potential applied, the electrical charges are accumulated at the dielectric layer, leading to reduction of the interfacial energy and thus of the contact angle between the dielectric material surface and the conductive liquid. By introducing the dielectric layer, electrical breakdown can be effectively evaded, facilitating control of meniscus shape with significantly lower voltage and power. Since many conductive liquids used in EWOD can differ in opacity and refractive indices, EWOD is widely applied to optical devices including displays [2, 3], lenses [4], and prisms [5-7].

To act as an optical surface, the liquid-fluid interface should satisfy multiple functional requirements. For dynamic operations of displays and optical beam guides, the interface must be quickly transformed to a desired shape with minimal overshoot. The interfacial waves generated during the rapid motion of the interface in the optofluidic devices were analyzed previously, where the importance of making the cell small enough for capillarity to dominate viscosity and inertia was stressed [8]. Once moved to the desired position and shape, the interface should be stably maintained. It is known that direct current (DC) is disadvantageous in that aspect because accumulation of counterions at the wall over a long period may lead to damage of the dielectric surface [9, 10]. As a remedy, AC (alternating current) with the magnitude of voltage corresponding to the desired contact angle has been suggested [10]. However, oscillations of the interface under AC conditions have been reported which are detrimental to the optofluidic system's performance.

Long-term durability of the EWOD-based system is enhanced by the right choice of electrolytes in conductive liquid as well as by the use of AC (as explained above) and high quality dielectric layer [11, 12]. It was reported that electrolytes with large polymer chains can increase the breakdown voltage as compared with small ions because the large chains are less likely to penetrate defects in the dielectric layer [10, 13-15].

Here we show that there exists a critical AC frequency over which the oscillation of the interface is prevented almost perfectly, whereas the interface oscillation is suddenly discernible at the frequency lower than the critical value. The frequency is determined by the duration when the contact angle is kept

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stable at each polarity condition of the wall. The stable duration is a sensitive function of the size of the charged matters in the solution.

In the following, we start with describing the experimental setup used to actuate three-phase contact lines by EWOD and to observe their stability and motions. We then present experimental results of the transient response of the contact line upon change of voltage, which reveal the effect of electrolyte type. We find a critical AC frequency over which interfacial oscillation is significantly suppressed, which is explained through quantitative measurement result of contact angle.

## 2. Experiments and analysis

### *2.1 Experimental setup*

To observe the fluid-fluid interface motions driven by electrowetting, we fabricated microcells filled with two immiscible liquids (one conductive and the other nonconductive) as shown in Fig. 1. Two parallel electrode plates of $w = 5$ mm in width, made of glass wafer coated with molybdenum (electrode) and dielectric layers of silicon nitride and parylene C, are separated by a gap $d = 300$ μm, a typical size of real EWOD-based optical devices [2, 16]. The side walls are transparent glass coated with PTFE (polytetrafluoroethylene) less than 50 μm in thickness. As a nonconductive liquid, we used a mixture of 99 wt% 1-chloronaphthalene (Acros) and 1 wt% n-dodecane (Sigma-Aldrich) for its high refractive index over 1.6 and transmittancy [17]. We employed two different aqueous solutions as a conductive liquid - 0.85 wt% PAA (polyacrylic acid, Sigma-Aldrich) and 0.85 wt% branched PEI (polyethyleneimine, Sigma-Aldrich). The density and viscosity of the nonconductive oil mixture (liquid L) are 1179 kg/m$^3$ and 0.0027 Pa·s. Those of the aqueous solutions of PAA and PEI are 1000 kg/m$^3$ and 0.0013 Pa·s and 1000 kg/m$^3$ and 0.0010 Pa·s, respectively. The interfacial tension between the oil mixture and the aqueous solutions, $\gamma$, is measured to be 7.64 mN/m by the du Noüy ring method (Kruss K100 tensiometer). All the properties are for the room temperature. The capillary length at which the capillary and gravitational effects are balanced, $l_c = \sqrt{\gamma / (\Delta\rho g)} = 2.0$ mm, where $\Delta\rho$ is the density difference of the two overlapping immiscible liquids

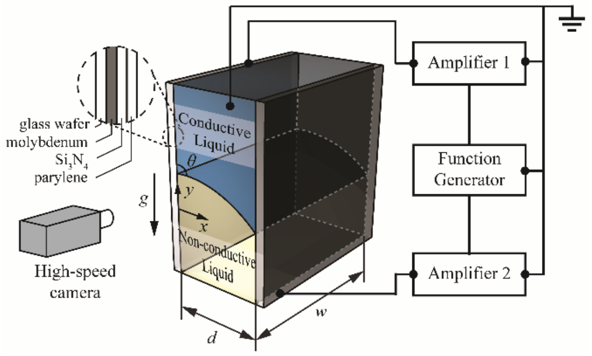

Fig. 1. Schematics of the microcell and the electrical setup to actuate the liquid-liquid interface by EWOD.

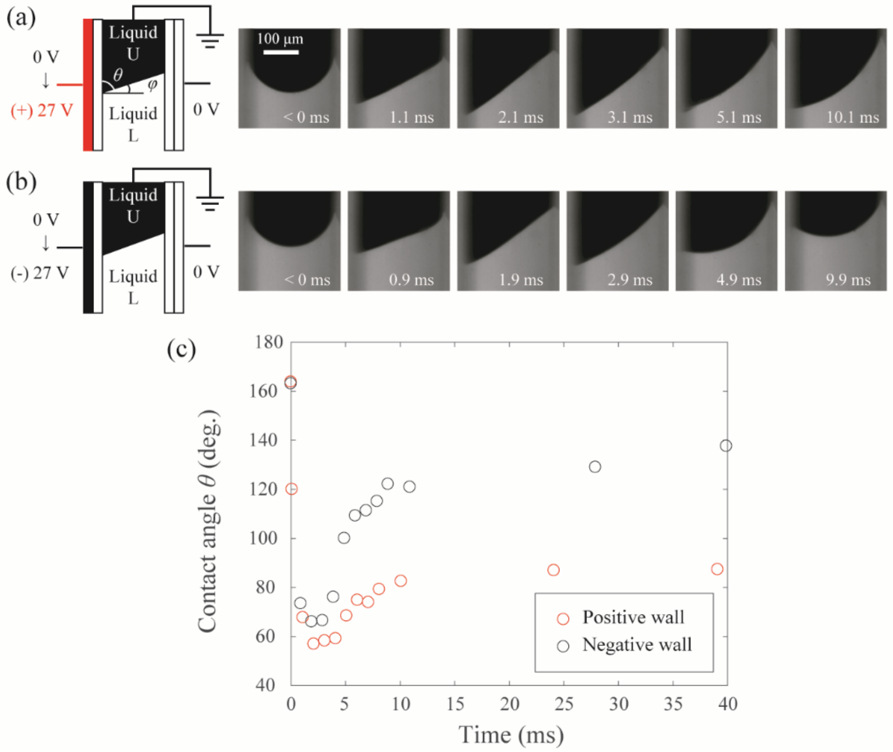

Fig. 2. Time series of the liquid-liquid interface motion when DC voltage is applied to the left wall. Meniscus shapes under the DC voltage of (a) +27 V; (b) -27 V; (c) measurement results of the contact angle.

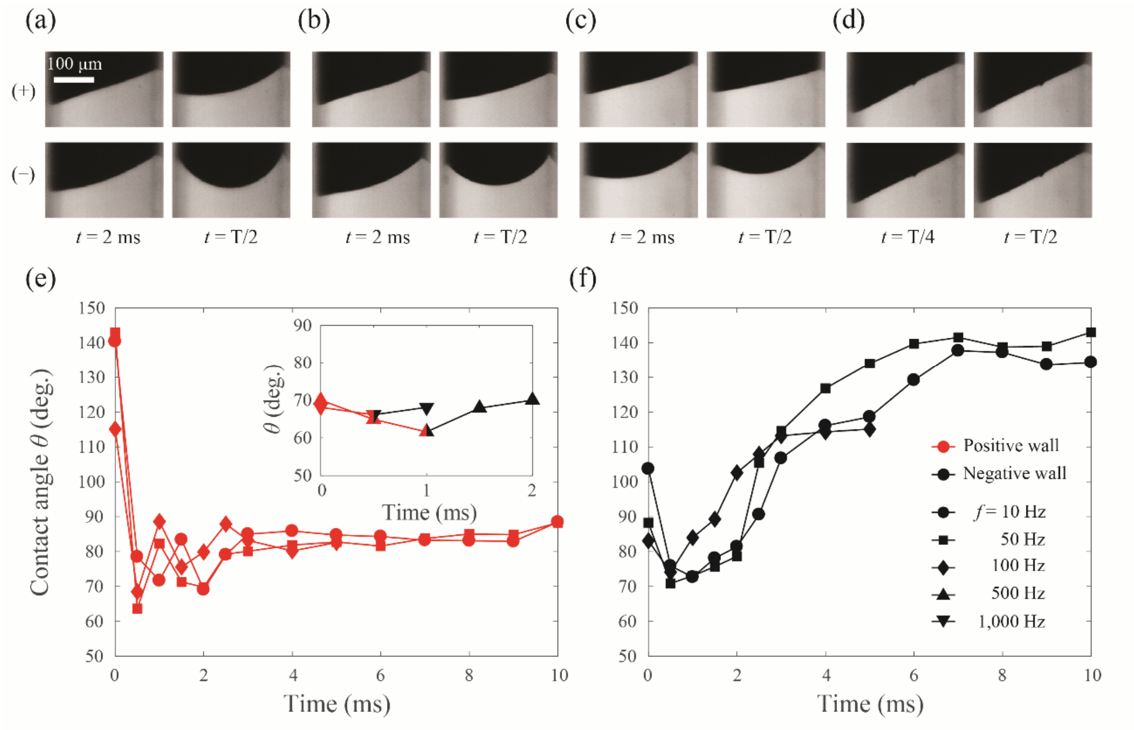

Fig. 3. Time series of the liquid-liquid interface motion in the EWOD cell when AC voltage is applied. Meniscus shapes for (a) $f$ = 10 Hz; (b) $f$ = 50 Hz; (c) $f$ = 100 Hz; (d) $f$ = 500 Hz. The polarity of wall voltages is indicated in (a). Measurement results of the contact angle when the polarity of the left wall is (e) positive; (f) negative. Inset: Contact angle for $f$ = 500 Hz (t = 0-2 ms) and 1000 Hz (t = 0-1 ms). The legends for (e) and (f) are given in (f).

and $g$ is the gravitational acceleration.

In the microcell with $d$ = 300 µm, which is significantly less than $l_c$ implying dominant effects of capillarity over gravity, the meniscus shape is determined by voltages of both the electrodes just as in a liquid prism [5, 6]. The electrodes are connected to high voltage amplifiers (AA Lab Systems A-301 HS) that amplify the driving voltage generated by a function generator (National Instruments NI9263). The function generator is controlled by a personal computer via a Labview (National Instruments) interface. The liquids are electrically grounded. Upon applying voltage either in DC or AC at a specified frequency, the motion of the interface is observed through the transparent side of the cell via a high-speed video camera (Photron APX-RS) recording images up to 10000 frames per second.

## 2.2 Experimental results

### 2.2.1 Response to DC

We first describe the dynamic response of the interface at the EWOD wall when the DC voltage is applied at time $t$ = 0. Figs. 2(a) and (b) show the image sequences of the interfaces between liquids U and L when +27 V and -27 V is applied to the left wall, respectively. We measured the corresponding contact angle of the interface with time, and the results are plotted in Fig. 2(c). Here the contact angle $\theta$ is measured from the solid wall through liquid U to the interface as depicted in Fig. 2(a).

When DC of +27 V is applied at $t$ = 0, the contact angle at the left wall abruptly drops from the initial value of 165° to approximately 60° within 2 ms as shown in Fig. 2(c). We designate the duration for the contact angle to adjust to the value corresponding to the EWOD voltage as $\tau$. When $t > \tau = 2$ ms, the contact angle rather increases or relaxes, indicating failure in keeping the liquid interface stable.

We see that the recovery of the contact angle is more severe for the negatively charged electrode. The dependency of the electrowetting response upon polarity of the EWOD wall can be attributed to the electrolyte infiltration in dielectric layer [10, 13-15]. The PAA solute is the polymeric electrolyte that is dissociated into large polymeric anions with many carboxyl units and small hydronium cations in water. Large anions are attracted toward the EWOD wall with the positive voltage, while small cations come closer to the wall with the negative voltage. The small positive ions permeate through the dielectric layer more easily, weakening the electric field of EWOD capacitor. Therefore, the regrowth of contact angle at negative DC voltage is more serious than at positive DC voltage.

A similar dependency of contact line response upon electrode polarity was observed with a different electrolyte solution of PEI. Since PEI is the polymeric electrolyte that is dissociated to large polymeric cations, with many ammonium units and small hydroxyl anions ($OH^-$) in water, the electrowetting response is more stable when negative voltage is applied. For both the electrolyte solutions, $\tau$ is 2 ms regardless of DC polarity.

### *2.2.2 Response to AC*

Upon testing the temporal stability of contact angle of each electrolyte solution depending on the electrode polarity, we applied square-wave AC voltage with various frequencies (10, 50, 100, 500 and 1000 Hz) to observe the effect of AC frequencies on the interface shape. The voltage magnitude applied to the left (21 V) and right (11 V) walls of the cell was such that it maintains the inclination of the liquid prism (designated in Fig. 2(a)) at $\varphi = 25°$. Here liquid U is the PAA solution. Figs. 3(a)-(d) show the temporal evolutions of the liquid interface shape when the actuation frequency, $f$, is 10, 50, 100 and 500 Hz. In each case, the constant voltage is maintained for $T/2$ with the period $T = 1/f$. In Figs. 3(a)-(c), the interface loses its stability within $T/2$ because $T/2 > \tau$. Also, we see that such relaxation is severer for the negative charged walls for the reason delineated above. Figs. 3(e) and (f), the measurement results for the contact angle for each polarity, quantitatively show the relaxation of the interfacial profiles for 10, 50 and 100 Hz.

When the frequency is further increased to 500 Hz, the interface shape is kept stable for the half period as shown in Fig. 3(d). Inset of Fig. 3(e) displays the behavior within the initial 2 ms in detail, revealing that the contact angle is maintained stable for the half-period for $f = 500$ and 1000 Hz regardless of the electrode polarity. The fluctuation of the contact angle, or the deviation of the contact angle from the average value, for 1 s is within ±6.4 % for 500 Hz and ±1.5 % for 1000 Hz. It is because the half period $T/2$ is shorter than the maximum duration of stable interface $\tau = 2$ ms. Dissociated ions do not have sufficient time to permeate the electrode wall, so that the reduced contact angle is maintained for each half period.

The foregoing observations lead us to conclude that the liquid interface operated by the AC EWOD scheme is stable when the AC frequency is such that $f > 1/(2\tau)$. Here $\tau$ is the duration for which the contact angle is reduced to the value corresponding to the EWOD voltage, a result of the DC EWOD test. Such time scale for the contact angle change is observed to hold for low frequency AC cases as well. When the frequency is increased to the level when the half period is shorter than $\tau$, the contact angle does not have sufficient time to relax and so is kept stable over time despite periodic changes of the electrode voltage.

## 3. Conclusions

Although the origin of the interfacial fluctuation under AC operation of EWOD devices and the frequency condition to eliminate such ripples have been identified in this work, the physical mechanism to determine the time scale $\tau$ needs further investigation. It is associated with the time scales not only for the contact angle to change from its value under neutral condition to the one under EWOD voltage, but also for the dissociated ions to permeate the electrode wall. An observation of the contact angle change with time reveals that although the magnitude of contact angle relaxation is greater on the wall with the polarity opposite to the charge of small ions (negative wall for PAA and positive wall for PEI), the time that the interfacial relaxation begins is similar regardless of the wall polarity. A detailed study on the physicochemical interaction of EWOD electrode materials with dissociated ions will help one to design and build stable yet fast EWOD devices for a variety of applications including optofluidics.

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## Nomenclature

| | |
|---|---|
| $d$ | : Gap between two EWOD plates |
| $f$ | : Frequency |
| $g$ | : Gravitational acceleration |
| $l_c$ | : Capillary length |
| $T$ | : Time period of AC voltage wave |
| $w$ | : EWOD cell width |
| $\gamma$ | : Interfacial tension of two immiscible liquids |
| $\Delta\rho$ | : Density difference of two immiscible liquids |
| $\theta$ | : Contact angle of conductive liquid |
| $\tau$ | : Maximum duration of stable interface |
| $\varphi$ | : Tilt angle of liquid prism |

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