Electrowetting on dielectric driven droplet resonance and mixing enhancement in parallel-plate configuration

Chiun-Peng Lee,^{1,2} Hsin-Chien Chen,^{1,2} and Mei-Feng Lai^{1,2,a)}

¹*Department of Power Mechanical Engineering, National Tsing Hua University, Hsinchu, Taiwan*

²*Institute of Nanoengineering and Microsystems, National Tsing Hua University, Hsinchu, Taiwan*

(Received 19 August 2011; accepted 8 December 2011; published online 15 March 2012)

This study experimentally verifies that the mixing process in a droplet can be enhanced by driving the droplet at resonant frequencies and at alternating driving frequencies using a parallel-plate electrowetting on dielectric device. The mixing time, which is defined as the time required for reaching the well-mixed state, in a resonant droplet is found to be significantly shorter than that in a non-resonant droplet. Besides, it is also found that a higher driving potential leads to a better mixing effect, especially at resonant frequencies. Furthermore, when a droplet is driven by alternating two driving frequencies, especially two resonant frequencies, the mixing efficiency is found to be significantly enhanced for a specific alternating duration of these two frequencies. © 2012 American Institute of Physics. [doi:10.1063/1.3673258]

I. INTRODUCTION

Due to the advantages of rapid response and the programmability of droplet movements, electrowetting on dielectric (EWOD) (Ref. 1) has become a major platform for driving droplets in microfluidic systems. Fundamentals of EWOD, including basic operations on a droplet,²⁻⁴ dynamics of contact angle variation,^{5,6} and contact angle saturation^{7,8} have been extensively studied. Integrated functional chips⁹⁻¹¹ and in-droplet particle manipulation^{12,13} based on EWOD have also been demonstrated. For applications using EWOD, especially in chemical and biochemical fields, efficient mixing of samples and reagents is always desired and is still an issue. The EWOD-based digital microfluidics is typically classified into two configurations: single-plate and parallel-plate EWOD. Comparing these two configurations, a single-plate EWOD device is easier to integrate with other devices, and the smaller liquid-solid contact area in a single-plate EWOD device allows the droplet to move faster.¹⁴ In terms of droplet splitting, separating smaller droplets from a bigger droplet can be easily achieved on a single-plate EWOD device by using the hydrodynamic instability of a long liquid column.¹⁵ The variation of species concentration, however, is an issue due to the higher evaporation rate in single-plate EWOD devices.³ On the other hand, when using a parallel-plate EWOD device, a smaller gap between two plates can significantly enhance the efficiency of droplet cutting.³ Therefore, the parallel-plate EWOD may be considered as an alternative for some applications. In general, mixing using parallel-plate EWOD actuation is slower than using single-plate EWOD actuation.¹⁴ Previous research¹⁶ shows that the mixing time required in parallel-plate EWOD actuation is found to be reduced by moving the droplet on a digitally controlled electrode array in specific loops, but this mixing mechanism may complicate the chip design. To achieve a more efficient mixing for droplet-based microfluidic applications using parallel-plate EWOD devices, mixing enhancement using droplet resonance is studied experimentally in this study. The mixing efficiency of a droplet under resonant oscillations and non-resonant oscillations is discussed to verify the influence of droplet resonance on the mixing.

^{a)}Electronic mail: mflai@mx.nthu.edu.tw. Tel: +886-3-5715131 ext. 33998. Fax: +886-3-5745454.

Up to now, most of the previous researches on EWOD-driven droplet oscillation mainly focuses on the single-plate EWOD actuation, including hydrodynamic flows in an oscillating droplet^{17,18} and the relationship between forcing frequency and mixing efficiency,¹⁹ the shapes of self-excited oscillating droplets,^{20,21} and the droplet oscillation spectrums and the beat phenomenon of droplet oscillation near the resonant frequency.²² In this study, we focus on the droplet oscillation in a parallel-plate EWOD device, and study the mixing enhancement caused by the droplet resonance. From our experimental results, the mixing time in a resonant droplet can be significantly reduced to $7.4\% \pm 0.7\%$ of the time needed for a non-oscillated droplet to reach the well-mixed state.

II. MATERIALS AND METHODS

The schematic illustration of the parallel-plate EWOD device in our experiment is shown in Fig. 1(a). Both of the top and bottom plates are of indium tin oxide (ITO) glass. The top substrate is the ground electrode and the bottom substrate is the driving electrode. The gap d between these two plates is set to be $700\ \mu\text{m}$. $5.6\ \mu\text{m}$ thick poly(methyl methacrylate) (PMMA) and $60\ \text{nm}$ thick Teflon AF (DuPont Corp.) are spin coated above the electrode to be the dielectric layer and hydrophobic layer, respectively. To excite the droplet resonance, the driving potential is a periodic square wave with offset as shown in Fig. 1(b). The droplet is actuated/relaxed during the active/inactive period. The time interval of the applied square wave voltage is indicated by a parameter T in Fig. 1(b).

The experimental setup of the frequency scanning system,²² which is used here to derive the resonant frequencies and the oscillation spectrum of a droplet, is shown in Fig. 2(a). The He-Ne laser (25LHP151-249, Melles Griot) is used as the probing beam incident from one side of droplet. When the droplet oscillates, the laser beam passing through the droplet oscillates accordingly. The position sensor (PSM2-10Q, On Trak) placed at the other side of droplet is used to measure the movement of the transmitted laser beam. The periodic driving potential is provided by a function generator (SFG830, GW INSTEK) connected to a high voltage amplifier (HA-800, Pintek). The signal voltage from the position sensor is monitored by a lock-in amplifier (SR830, Stanford Research Systems), which is phase locked to a reference frequency provided by the function generator. We measure the relative amplitudes of droplet oscillation at different driving frequencies.

Fig. 2(b) shows the experimental setup for the droplet mixing experiment. The $7\ \mu\text{l}$ merged droplet consists of two colored droplets both with volume $3.5\ \mu\text{l}$ is placed on the substrate. The colorings in our experiments are red and green edible pigments. The volume concentration of the pigment for each red and green deionized water droplet is about 1.2%. The $7\ \mu\text{l}$ droplet size we chose is close to the droplet sizes adopted in some relevant studies about EWOD in parallel plate configurations.^{9,14} For our study, the droplet aspect ratio (width to height) is about 5.1, and the ratio of solid-liquid interface to the liquid-vapor interface is about 2.55 under a simple assumption that the droplet shape is a short cylinder. These ratios can also be determined when the gap is $0.1\ \text{mm}$ and the droplet volume is $80\ \text{nL}$, which is very similar to the device used in a

FIG. 1. Schematic plots for the device and driving potential in the experiment. (a) The device for parallel-plate EWOD. (b) Driving electric potential in a periodic square waves with offset. The droplet is actuated/relaxed during the active/inactive period.

FIG. 2. Schematic plots for the experimental setup. (a) The frequency scanning system. (b) Setup for the droplet mixing experiment.

previous literature.¹⁰ When the periodic driving potential is applied, the images of the mixed droplet are taken by an optical microscope equipped with a charge-coupled device (CCD) camera. In this experiment, a snapshot of the droplet is taken for every 5 s during the mixing, and we calculate the degree of mixing by analyzing the hue distribution within the droplet. A JAVA-based image processing software IMAGEJ is used to analyze the hue of pixels.

III. RESULTS AND DISCUSSION

Figure 3 shows the average amplitude spectrum for the oscillating water droplet in the parallel-plate EWOD. The amplitude of the voltage output from the position sensor is proportional to the amplitude of the water droplet oscillation. Figure 3 is the average data of five measurements. The major peaks in Fig. 3 correspond to the droplet's resonant frequencies at different resonant states, which are 34, 66, 99, 131, and 165 Hz. To verify the influence of the droplet resonance on the mixing, the droplet is also oscillated at non-resonant frequencies. In our experiments, the errors on the measured droplet's resonant frequencies 34, 66, 99, 131, and 165 Hz are all within the range $\pm 1 \sim \pm 2$ Hz. From the spectrum in Fig. 3, it can be observed that the major peaks corresponding to the maxima of the droplet's oscillating amplitude are quite broad, which means that one or two Hz error on the measured droplet's resonant frequencies causes little influence on droplet's oscillating amplitude. In other words, the $\pm 1 \sim \pm 2$ Hz errors on the measured droplet's resonant frequencies can be considered to make a little impact on the mixing time, which is strongly correlated to droplet's oscillating amplitude.

To quantitatively analyze the droplet mixing, we define a mixing index η_k to specify the degree of mixing for the k th state

FIG. 3. The average amplitude spectrum for a $7 \mu\text{l}$ water droplet in parallel-plate EWOD. The average amplitude for each driving frequency is obtained from 500 measured data points within 5 s, and error bars indicate the standard deviation of oscillation amplitudes.

$$\eta_k = \frac{\phi_k - \phi_i}{\phi_f - \phi_i}, \quad (1)$$

where ϕ_i , ϕ_f , and ϕ_k denote the standard deviation of hue distribution within the droplet at the initial state, the completely mixed state, and the k th state during the mixing, respectively. Figures 4(b) and 4(d) show the experimentally obtained hue spectrum of the droplet at the initial and the completely mixed state, respectively, without EWOD actuation, where the mixing only depends on diffusion. The hue spectrum shown in Fig. 4(d) is the reference for the following mixing experiments using EWOD actuation. Figures 4(a) and 4(c) show the pictures of the droplet before and after mixing, respectively. The experimentally obtained time-dependent mixing index variation in the absence of EWOD actuation is shown in Fig. 5. The initial state and the completely mixed state correspond to $\eta = 0$ and $\eta = 1$ in this figure. We define $\eta = 0.95$ as the well-mixed state, where 95% of the mixing is completed, and the time required for reaching the well-mixed state is defined as mixing time. The mixing time for a droplet without EWOD actuation is 673 ± 7 s.

Figure 6 shows the average mixing time when the droplet is driven at different driving frequencies and driving voltages. The droplet is driven at resonant frequencies, 34, 66, 99, 131, and 165 Hz, and two non-resonant frequencies, 83 and 147 Hz, with 180 V and 300 V peak-to-peak driving potentials. In this figure, the mixing times indicated by bars are the averaged value of 3 measurements. The top and bottom values of error bars indicate the maximum and the minimum mixing times, respectively. From Fig. 6, comparing to the case without EWOD actuation (0 Hz), it can be observed that the mixing time is significantly reduced when exciting the droplet resonance. For example, when the droplet is driven by 300 V periodic driving potential at the resonant frequency 165 Hz, the mixing time required to reach $\eta = 0.95$ is 50 ± 5 s, which is only $7.4\% \pm 0.7\%$ of the time needed for a non-oscillated droplet to reach the well-mixed

FIG. 4. (a) The top view of the droplet before mixing. (b) The numbers of pixels versus different hues at the initial state before mixing. (c) The top view of the droplet after mixing. (d) The numbers of pixels with different hues at the completely mixed state.

FIG. 5. Time-dependent mixing index variation in the absence of EWOD actuation.

state. In contrast, when the droplet is driven at its non-resonant frequencies, the mixing times become much longer, though they are still much shorter than the mixing time for a non-oscillated droplet. From previous research^{19,23–25} that as the droplet oscillates, steady circulations are induced inside the droplet by the periodic deformation of the droplet surface. The significant reduction of the mixing time of the oscillating droplet in our experiment is believed to come from the enhanced mixing due to the forced convection. This effect is relatively stronger when the droplet is excited at its resonant frequencies, at which the droplet oscillating amplitudes increase dramatically such that the induced convection inside the droplet becomes very large.

For a droplet oscillated by EWOD actuation, applying a larger driving potential can produce larger variation of the Young's contact angle, that is, more pronounced displacement of the three-phase contact line.^{2,7,26,27} It infers that at the same driving frequency, a greater droplet deformation can be excited by actuating the droplet with a larger alternating driving electric signal. For the electric driving potential used in this study (see Fig. 1(b)), 300 peak-to-peak driving voltage can produce larger droplet oscillation amplitude than 180 V. Therefore, similar to the comparison of the resonant and non-resonant cases, stronger internal flows can be induced by applying the driving potential 300 V. In consequence, the mixing in the cases of using 300 V driving potential are faster than the cases of using 180 V (see Fig. 6), especially when the droplet is driven at its resonant frequencies.

The mixing performance depends not only on the droplet's oscillation amplitude but also on the distribution of the internal flows. Different flow patterns can be induced by exciting the droplet resonance at different resonant frequencies, that is, different resonant states of the droplet that are determined by the number of nodes on the vapor-liquid interface.^{17,24,28–31} Therefore, although the droplet's oscillation amplitude decreases progressively from 34 Hz driving frequency

FIG. 6. The average mixing time required for reaching the well-mixed state for the droplet driven at different driving frequencies and driving voltages.

to 165 Hz driving frequency (see Fig. 3), the fastest mixing occurs at 165 Hz instead of 34 Hz. In Fig. 6, when increasing the driving frequency from 34 Hz to 99 Hz, the figure shows an increase in the mixing time, or a decrease in efficiency, which could be explained by the fact that the amplitudes of droplet oscillation are decreasing. The mixing time peaks after 99 Hz decrease to values even lower than at 34 Hz. This could be explained by the different flow profiles within the droplet. Figure 7 shows the photos of color distributions in a droplet during the mixing at 34, 99, and 165 Hz. In this figure, quite different mixing processes at 165 Hz can be found. The rotation of color distribution can only be observed at 165 Hz, and it is believed to be the reason why the mixing performance is higher at 165 Hz. Another reasonable explanation for the relationship between the mixing efficiency in a small oscillating droplet and the driving frequency was proposed on the concept of time scales.³² If the time scales of the driving frequency and the time required of a fluid particle to travel along whole flow path of a closed streamline are close, the mixing would be dramatically enhanced. This supports the rationality of our finding that different droplet resonant states cause different mixing enhancements.

In this study, the influence of alternating resonant states on droplet mixing is also verified. From previous research, it is known that the internal flow fields vary inside the droplet for different droplet resonant states. Therefore, it is expected that the alternating internal flow fields may affect the mixing effect. To emphasize the advantage of the mixing method of using two alternating frequencies, we adopted two frequencies that correspond to longer mixing times to verify the pronounced mixing enhancement. When the droplet is driven with 180 V driving potential between two alternating resonant frequencies 34 Hz and 99 Hz, each of which lasts for 5 s, the average time required to reach the well-mixed state ($\eta = 0.95$) can be reduced to 127 s (see Fig. 8), which is far below the mixing time 190 s for 34 Hz and 228 s for 99 Hz as shown in Fig. 6. Similarly, the mixing time can also be reduced when the droplet is driven between two alternating non-resonant frequencies 83 Hz and 147 Hz (see Figs. 6 and 8). This implies that to break and reconstruct streamlines periodically can produce additional chaos to enhance the mixing further. Comparing the resonant and non-resonant cases, we find that applying the alternative manner with strong flows induced by droplet resonance provides a relatively greater opportunity to improve the mixing. Note that in Fig. 8, 3 s, 5 s, 10 s, and 20 s are adopted to test the influence of time durations on the mixing efficiency. Considering the time required for the droplet to reach the steady state after applying a periodic driving voltage, 3 s can be reasonably chosen as the shortest duration for alternating two frequencies to ensure that the droplet can oscillate in a steady state at both frequencies. The lowest adopted duration 3 s is also the limitation of our instrument. Interestingly, in Fig. 8, for both resonant and non-resonant cases,

FIG. 7. Sequential images of the droplet mixing process. The driving frequency and driving potential are 34 Hz and 180 V_{pp} for (a), 99 Hz and 180 V_{pp} for (b), and 165 Hz and 300 V_{pp} for (c).

FIG. 8. The average mixing time required for reaching the well-mixed state when the droplet is driven between two alternating resonant frequencies 34 and 99 Hz and two alternating non-resonant frequencies 83 and 147 Hz. The peak-to-peak driving voltage is 180 V.

the 5 s alternating duration is found to be the optimum choice for the rapid mixing when the droplet is oscillated by alternating two driving frequencies. These results show that the durations of the two alternating driving frequencies significantly influence the mixing process, and the mixing efficiency is found to be significantly enhanced for a specific alternating duration of these two frequencies.

IV. CONCLUSIONS

In conclusion, enhancement of the mixing process by driving the droplet at its resonant frequencies and by applying alternating driving frequencies are verified experimentally in parallel-plate EWOD devices. The experimental results verify that a resonant droplet has higher mixing efficiency than a non-resonant droplet. Alternating the internal flow with a proper period is also found helpful for the mixing enhancement. Using droplet resonance to produce strong internal flows and intervening the streamlines to produce chaos for accelerating the chemical process are promising in the applications of biochips and EWOD-based microfluidics. Besides, our mixing approach does not need the electrode array, and this can significantly simplify the device design and control circuit. This is very beneficial for integrating the device with others to construct a fully functional biochip to perform complex biology studies.

ACKNOWLEDGMENTS

This work was supported partly by the ROC National Science Council Grant Nos. NSC 99-2112 -M-007-016-MY3 and NSC 99-2112 -M-007-015-MY3.

- ¹B. Berge, C. R. Acad. Sci. II **317**, 157 (1993).
- ²M. G. Pollack, A. D. Shenderov, and R. B. Fair, *Lab Chip* **2**, 96 (2002).
- ³S. K. Cho, H. Moon, and C. J. Kim, *J. Microelectromech. Syst.* **12**, 70 (2003).
- ⁴M. Abdelgawad, S. L. S. Freire, H. Yang, and A. R. Wheeler, *Lab Chip* **8**, 672 (2008).
- ⁵K. H. Kang, *Langmuir* **18**, 10318 (2002).
- ⁶J. Berthier, Ph. Clementz, O. Raccurt, D. Jary, P. Claustre, C. Peponnet, and Y. Fouillet, *Sens. Actuators, A* **127**, 283 (2006).
- ⁷K. Adamiak, *Microfluid. Nanofluid.* **2**, 471 (2006).
- ⁸A. I. Drygiannakis, A. G. Papathanasiou, and A. G. Boudouvis, *Langmuir* **25**, 147 (2009).
- ⁹V. Srinivasan, V. K. Pamula, and R. B. Fair, *Lab Chip* **4**, 310 (2004).
- ¹⁰H. Moon, A. R. Wheeler, R. L. Garrell, J. A. Loo, and C. J. Kim, *Lab Chip* **6**, 1213 (2006).
- ¹¹L. Malic, T. Veres, and M. Tabrizian, *Lab Chip* **9**, 473 (2009).
- ¹²S. K. Cho, Y. Zhao, and C. J. Kim, *Lab Chip* **7**, 490 (2007).

- ¹³G. J. Shah and C. J. Kim, [Lab Chip](#) **9**, 2402 (2009).
- ¹⁴C. G. Cooney, C. Y. Chen, M. R. Emerling, A. Nadim, and J. D. Sterling, [Microfluid. Nanofluid.](#) **2**, 435 (2006).
- ¹⁵T. B. Jones, M. Gunji, M. Washizu, and M. J. Feldman, [J. Appl. Phys.](#) **89**, 1441 (2001).
- ¹⁶Y. H. Chang, G. B. Lee, F. C. Huang, Y. Y. Chen, and J. L. Lin, [Biomed. Microdevices](#) **8**, 215 (2006).
- ¹⁷S. H. Ko, H. Lee, and K. H. Kang, [Langmuir](#) **24**, 1094 (2008).
- ¹⁸H. W. Lu, F. Bottausci, J. D. Fowler, A. L. Bertozzi, C. Meinhardt, and C. J. Kim, [Lab Chip](#) **8**, 456 (2008).
- ¹⁹F. Mugele, J. C. Baret, and D. Steinhäuser, [Appl. Phys. Lett.](#) **88**, 204106 (2006).
- ²⁰J. C. Baret and M. M. J. Decré, [Langmuir](#) **23**, 5173 (2007).
- ²¹J. M. Oh, S. H. Ko, and K. H. Kang, [Langmuir](#) **24**, 8379 (2008).
- ²²M. F. Lai, C. P. Lee, C. N. Liao, and Z. H. Wei, [Appl. Phys. Lett.](#) **94**, 154102 (2009).
- ²³A. Bertelsen, [J. Fluid Mech.](#) **64**, 589 (1974).
- ²⁴E. Trinh and T. G. Wang, [J. Fluid Mech.](#) **122**, 315 (1982).
- ²⁵E. J. Chang and M. R. Maxey, [J. Fluid Mech.](#) **277**, 347 (1994).
- ²⁶J. H. Lee, H. Moon, J. Fowler, T. Schoellhammer, and C. J. Kim, [Sens. Actuators, A](#) **95**, 259 (2002).
- ²⁷J. Berthier, P. Dubois, P. Clementz, P. Claustre, C. Peponnet, and Y. Fouillet, [Sens. Actuators, A](#) **134**, 471 (2007).
- ²⁸E. Trinh, A. Zwern, and T. G. Wang, [J. Fluid Mech.](#) **115**, 453 (1982).
- ²⁹T. Watanabe, [Comput. Fluids](#) **37**, 91 (2008).
- ³⁰S. Oberti, A. Neild, R. Quach, and J. Dual, [Ultrasonics](#) **49**, 47 (2009).
- ³¹R. Malk, Y. Fouillet, and L. Davoust, [Procedia Chem.](#) **1**, 1107 (2009).
- ³²S. M. Lee, D. J. Kim, and I. S. Kang, [Phys. Fluids](#) **12**, 1899 (2000).