Droplet Ejection and Liquid-solid Separation from Covered Microfluidic Systems

Weiqiang Wang[1,2,*], **Thomas B. Jones**[2]

[1] School of Mechanical Engineering, Nanjing University of Science and Technology, Nanjing 210094, China
[2] Department of Electrical and Computer Engineering, University of Rochester, Rochester, New York 14627, USA
* Corresponding author: wangweiqiang@njust.edu.cn

Abstract In droplet-based microfluidics, two different types of microsystems are used for droplet manipulation - covered systems based on parallel-plate devices, and open systems based on single-plate devices. Ejecting droplets from covered systems into an open system is an essential operation to build a complete analysis system combining the advantages of both covered and open systems. This paper presents theoretical study and experimental results for ejecting a water and/or oil droplet from a covered into an open system. We use a force analysis method to determine the condition required for a droplet to cross the open/covered edge and demonstrate droplet movements using electric actuation. A bevelled edge structure is developed to facilitate the final liquid-solid separation process, greatly reducing the required actuation voltage.

Keywords electrowetting, electrowetting-on-dielectric (EWOD), microfluidics, dielectrophoresis

---

1. Introduction

Droplet-based microfluidics (or digital microfluidics) is a fast-developing liquid-handling technology that manipulates liquids in the form of discrete droplets. In integrated devices for droplet-based microfluidics, liquid droplets in contact with dielectric surfaces are moved by applying AC or DC potentials across electrodes patterned beneath the dielectric. There are two types of microsystems for electric actuation of liquid droplets: the covered system, in which voltages are applied across electrodes in both the top and bottom plates, [1-5] and the open system, in which voltages are applied across electrodes on a single plate. [6-10] The mechanism for droplet actuation has been attributed to electrowetting (EW) forces for conductive liquids [1-12], and to dielectrophoretic (DEP) forces for dielectric liquids. [13-16] When voltage is applied across electrodes in a device, a liquid droplet can move towards the activated electrode.

Study on droplet manipulation is usually done in either covered microsystems or open microsystems. The covered system is capable of achieving all the basic fluidic operations- droplet dispensing [3-5,18], moving [1,2], splitting [3,5] and merging [3], and controls droplet volume reliably through dispensing. [4,18] The open system, while lacking the capacity of dispensing or splitting droplets, [5] allows direct access to liquid handling tools on the surface and analytical equipments. Thus, the concept of dual open/covered microsystem combining the advantages of both covered and open systems is developed. [20,21] A covered section dispenses liquid droplets with desired volume and further analytical operation or liquid droplet handling is done on an open section. To achieve this scheme, it is essential to eject a droplet from a covered into an open section of a chip. In this paper, we analyze the possibility of such a motion for both water and oil droplets, and demonstrate experimental results to compare with the results of modeling. A bevelled edge on the top plate makes it possible to minimize the contact area between droplet and the top plate, thus easily separating the liquid-solid interface.

2. Water droplet ejection

An integrated device consisting of a covered section and an open section is illustrated in top and side views in Figure 1 (a) and (b). The bottom plate consists of a patterned array of addressable electrodes, and the top plate in the covered section is coated with a transparent indium tin oxide (ITO) layer as a continuous ground electrode. The electrodes on bottom plate are coated with a dielectric layer, and both the top and bottom surfaces are covered with Teflon-AF as a hydrophobic film.

To evaluate the prospect of droplet movement from covered to open section by electrical actuation, we conduct force analyses to compare the driving forces with resisting forces. These results provide an estimate for the required voltage for a droplet to achieve such a motion.

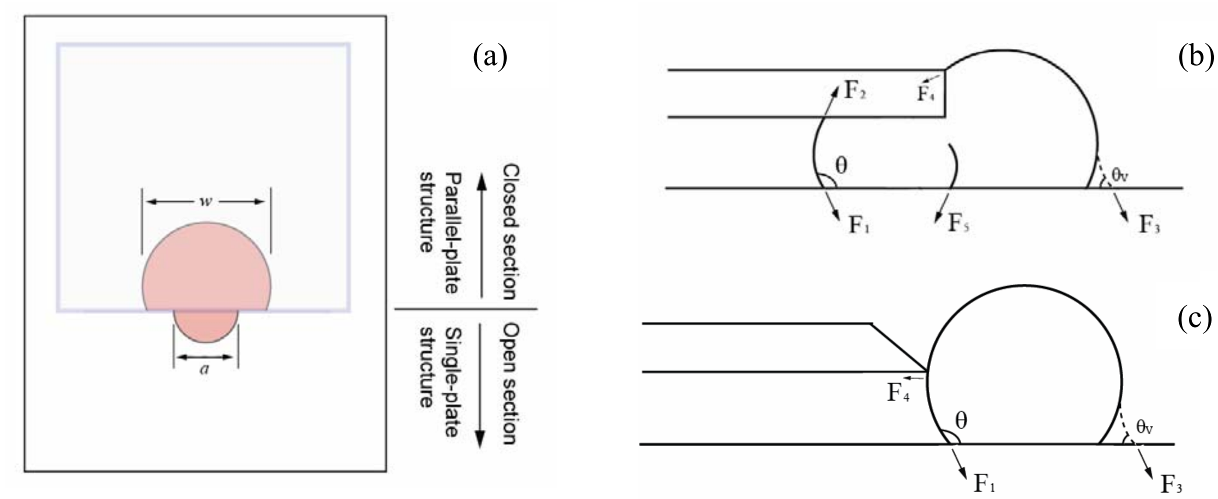

Figure 1. Force balance analysis of a water droplet ejection from covered into an open section. (a) Top view of a water droplet at the covered/open boundary. (b) Forces exerted on a water droplet when it is being moved toward the open section. (c) Liquid-solid separation with beveled edge.

2.1. Force balance analysis

The forces exerted on the droplet at the covered/open interface are illustrated in Figure 1 (b). We can distinguish two distinct contributions in the plane of the structure: the capillary force and the EW force. The capillary force is

$$F_c = F_1\left|\cos\theta\right| + F_2\left|\cos\theta\right| \tag{1}$$

and the EW force is

$$F_{EWOD} = F_3\left|\cos\theta_V\right| + (F_1 - F_5)\left|\cos\theta\right| \tag{2}$$

where $F_1 = F_2 = \gamma w$, $F_3 = \gamma a$, $F_5 = \gamma(w-a)$. w and a are contact lengths of the droplet in covered and open sections respectively, γ is the surface tension of the liquid droplet. θ_V and θ are the droplet contact angle with and without applied voltage.

When the water droplet is being moved from the covered section to the open substrate, the influence of F_4 can be neglected as there is no motion for the upper portion of droplet that contacts the edge of the ITO plate. Thus, the total force acting on the drop is:

$$F_{total} = F_3\left|\cos\theta_V\right| + (F_1 - F_5)\left|\cos\theta\right| + F_2\left|\cos\theta\right| \tag{3}$$

The three force components in equation (3) tend to move the droplet toward the open section, so we may expect that the droplet gets out the covered section easily. In fact, the water droplet tends to move out even without external applied voltage, in which case the net force becomes $F_2|\cos\theta|$.

When the droplet emerges from the covered section, it still tends to stick to the Teflon-coated ITO plate due to the low interfacial tension between water and Teflon. To facilitate the separation of the liquid droplet and the top plate, we bevel the edge of the top plate as shown in Figure 1 (c). Here the total force acting on the droplet is

$$F_{total} = (F_1 \left|\cos\theta\right| + F_3 \left|\cos\theta_V\right|) - F_4 = \gamma a(\left|\cos\theta\right| + \left|\cos\theta_V\right|) - \gamma a \tag{4}$$

Using $\theta = 115°$, $F_{total} > 0$ gives $\theta_V < 55°$. Because of the contact angle saturation effect, it is not easy to reduce the contact angle of a water droplet to lower than 55° by EW actuation. However, the electromechanical approach of EW theory shows that the EW force experienced by the droplet is independent of the surface profile. [13,14] This result indicates that contact angle saturation does not necessarily limit the maximum EW force that can be applied to a droplet. [17] Converting the contact angle to applied electric voltage by the Lippmann-Young equation, we get $V > 105V$ as the voltage requirement for liquid-solid separation.

2.2. Experimental results of water droplet ejection

We fabricate a number of covered/open integrated devices on glass substrates to test droplet ejection motion. The bottom substrate consists of aluminum electrodes, a 0.5 μm spin-on-glass (SOG) dielectric layer, and a 1μm Teflon-AF layer. The top substrate is coated with an ITO layer and also a 1μm Teflon-AF layer. The spacing between top and bottom substrate is set to be 100 μm.

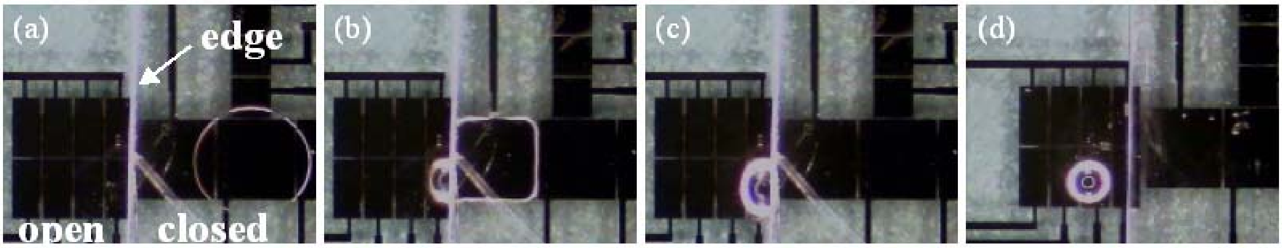

Figure 2. Moving a water drop from covered to open section. (a) The water droplet is initially placed in covered section. (b) The droplet is moved towards covered/open boundary by EW actuation using 100 V_{rms} 100 Hz AC. (c) The droplet enters open section as soon as it arrives at the boundary. (d) The droplet is successfully separated from top plate after applying 300 V_{rms} voltage.

Figure 2 shows video frames of water droplet motion from the covered to the open section. A water droplet is first injected into the covered section, and moved toward the covered/open interface by EW actuation. When the droplet reaches the interface, it is ejected rapidly but still sticks to the top plate. See Figure 2 (c). We then apply voltages ranging from 100 to 300 V_{rms} to separate the droplet and the top ITO plate. Separation is only successful at 300 V_{rms}.

To lower the required voltage for droplet/top plate separation, we bevel the edge of the top ITO plate to minimize the contact area between water droplet and Teflon surface. See Figure 3. Now the droplet can be easily separated by applying a voltage of 90 V_{rms}, which is even lower than the predicted value of 105 V. This is probably because the direction of force F_4 in Figure 1 (c) is not exactly horizontal, making the separation voltage somewhat lower.

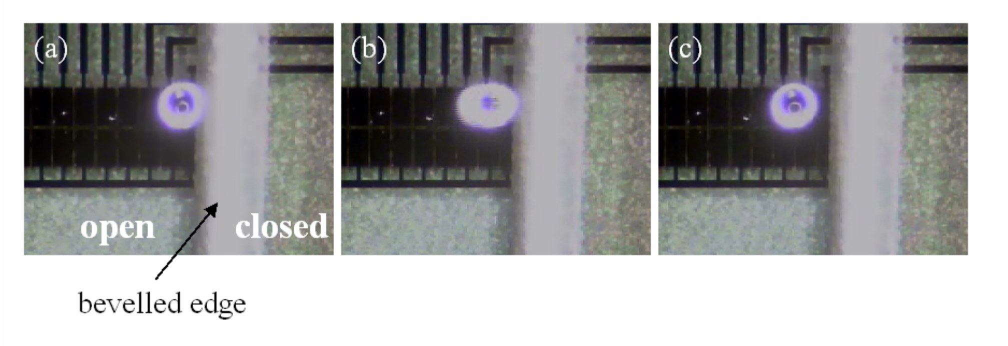

Figure 3. Separating a water drop and ITO plate with bevelled edges. The voltage applied is 90 V_{rms} 100 Hz AC.

3. Oil droplet ejection

Oil droplets have low surface tension and usually small contact angles on solid surfaces. It is likely that an oil droplet will find its way into cracks or small channels due to the capillary effect. As a consequence, it is very difficult to eject an oil droplet from covered into an open section.

3.1. Force balance analysis

The tendency of a liquid to enter narrow channels is commonly recognized as the capillary effect. A classic textbook example is the liquid rising in a vertical glass tube as shown in Figure 4 (a). For our apparatus, the liquid droplet moves horizontally and the goal is to use electrical actuation to overcome the capillary effect. To evaluate the requirements to accomplish this operation, we conduct a force analysis to compare the driving force and resisting forces as shown in Figure 4 (b). The total force acting on the oil drop is:

$$F_{total} = F_{DEP} - F_2 \cos\theta - F_4 \sin\theta + (F_3 \cos\theta + F_5 \cos\theta - F_1 \cos\theta)$$

$$\approx F_{DEP} - F_2 \cos\theta - F_4 \sin\theta \qquad (5)$$

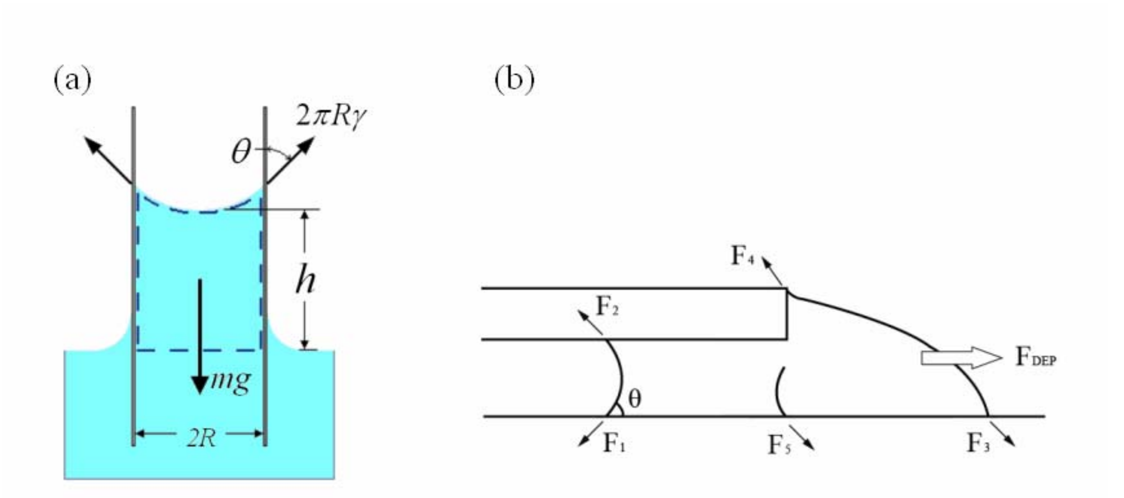

Figure 4. (a) Force analysis for a capillary tube. (b) Force analysis of an oil droplet at the covered/open boundary.

According to our device setup, [18,19] the minimum required voltage to achieve $F_{total} > 0$ is 1292 V and 1555 V for silicone oil and mineral oil, respectively. These high voltages are much greater than the dielectric breakdown voltages of 750 V, so it is impossible to achieve the desired movement by direct actuation. Furthermore, the oil droplet tends to get separated even the DEP force is large enough to overcome the resisting forces. We can understand this issue by considering a quasi-static process where the two plates are slowly separated as shown in Figure 5. The oil droplet sandwiched between parallel plates has a contact angle $\theta < 90°$. The vertical component of the surface tension forces at the upper (or lower) edge and the middle of drop are respectively:

$$F_{edge} = \gamma 2\pi R \sin\theta \tag{6}$$

$$F_m = \gamma 2\pi r \tag{7}$$

As the plates separate, both R and r decrease. Gradually F_{edge} will exceed F_m because r can approach 0. Then, the droplet breaks into two droplets, one each on the top and bottom substrates.

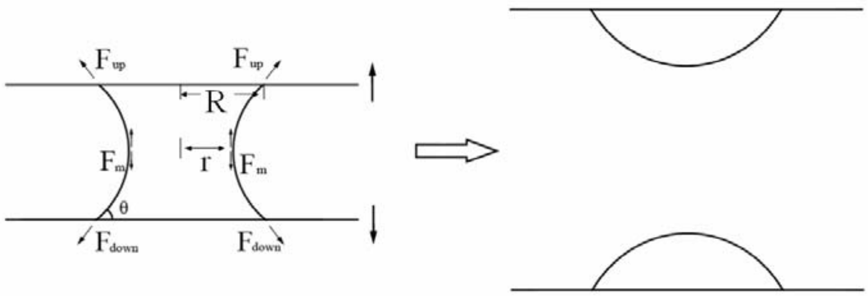

Figure 5. Breaking up of the oil droplet in parallel-plate structure when the two plates are separated far enough from each other.

A solution to this problem is to employ an oleophobic surface for the top plate. When the contact angle is > 90°, R at the upper edge is smaller than r, the oil droplet can be ejected without separation. And the total force acting on the oil droplet become

$$F_{total} = F_{DEP} + F_2 \left|\cos\theta^*\right| - F_4 \sin\theta \tag{8}$$

where θ^* is the oil contact angle against top surface, $\theta^* > 90°$. As F_2 and F_4 counterbalance each other mostly, the oil droplet can be ejected into an open section by DEP actuation.

3.2. Experimental results of oil droplet ejection

We fabricate an oleophobic surface on textured Si wafer and use it as the top plate for oil droplet ejection tests. The fabrication process of oleophobic Si microstructures is similar to that of Tuteja [22] and of Wu [23]. Figure 6 shows a mineral oil droplet sandwiched between a Teflon coated glass substrate (bottom) and a textured Si substrate (top) actuated by DEP. When the oil droplet is delivered to the covered/open boundary, continuous actuation moves the oil droplet to the open section, see Figure 6 (c).

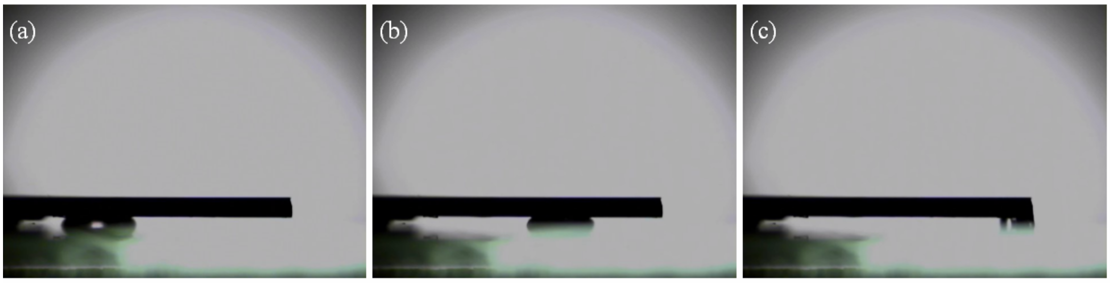

Figure 6. Side view showing the ejection of a mineral oil droplet. The spacing between Teflon coated glass substrate (bottom) and textured Si substrate (top) is 200 μm. The applied voltage is 560 Vrms 100 Hz AC.

The ejected oil droplet on open surface also sticks to the top plate. A beveled edge on the top plate may facilitate the detachment of the oil droplet. Further effort to fabricate an oleophobic beveled structure on the top plate is needed.

4. Conclusion

In this paper, we present theoretical predictions and experimental results on droplet ejection from covered into open microfluidic systems. We use a force balance analysis to predict the condition required to achieve such a motion. This model shows that it is easy to move a water droplet from covered to open section, but oil droplets always tend to stay in a covered section. Ejecting an oil droplet by direct DEP actuation is impossible on Teflon coated surfaces. An oleophobic surface must be used to complete this movement for oil droplet.

Experimental testing of the various predictions is done to demonstrate droplet ejection movements. To detach the water droplet from the top plate, we use a bevelled edge to minimize water-Teflon contact area and thus reduce the applied voltage. For oil droplets, we successfully use an oleophobic Si structure to achieve covered to open movement. Further study to bevel the Si structure is needed.

Acknowledgements

This work was supported by the U.S. Department of Energy, Office of Inertial Confinement Fusion under Cooperative Agreement No. DE-FC52-08NA28302, the University of Rochester, and the New York State Energy Research and Development Authority.

References

[1] M.G. Pollack, R.B. Fair, A.D. Shenderov, Electrowetting-based actuation of liquid droplets for microfluidic applications. Appl Phys Lett, 77 (2000) 1725–1727.
[2] M.G. Pollack, A.D. Shendorov, R.B. Fair, Electro-wetting-based actuation of droplets for integrated microfluidics. Lab Chip, 2 (2002) 96–101.
[3] S.K. Cho, H. Moon, C.J. Kim, Creating, transporting, cutting, and merging liquid droplets by electrowetting-based actuation for digital microfluidic circuits, J Microelectromech Syst, 12 (2003) 70-80.
[4] H. Ren, R.B. Fair, M.G. Pollack, Automated on-chip droplet dispensing with volume control by electro-wetting actuation and capacitance metering. Sens Actuators B, 98 (2004) 319–327.
[5] J. Berthier, Ph. Clementz, O. Raccurt, D. Jary, P. Claustre, C. Peponnet, Y. Fouillet, Computer aided design of an EWOD microdevice. Sens Actuators A, 127 (2006) 283–294.

[6] R.B. Fair, A. Khlystov, V. Srinivasan, V.K. Pamula, K.N. Weaver, Integrated chemical/biochemical sample collection, pre-concentration, and analysis on a digital microfluidic lab-on-a-chip platform. Proc of SPIE, 5591 (2004) 113-124.

[7] C.G. Cooney, C.Y. Chen, M.R. Emerling, A. Nadim, J.D. Sterling, Electrowetting droplet microfluidics on a single planar surface, Microfluid Nanofluid, 2 (2006) 435-446.

[8] M. Abdelgawad, A.R. Wheeler, Rapid prototyping in copper substrates for digital microfluidics, Adv Mater, 19 (2007) 133-137.

[9] P.Y. Paik, V.K. Pamula, K. Chakrabarty, Adaptive cooling of integrated circuits using digital microfluidics, IEEE Tran Very Large Scale Integr (VLSI) Syst, 16 (2008) 432-443.

[10] U.C. Yi, C.J. Kim, Characterization of electrowetting actuation on addressable single-side coplanar electrodes, J Micromech Microeng, 16 (2006) 2053-2059.

[11] F. Mugele, J.C. Baret, Electrowetting: from basics to applications, J Phys: Condens Matter, 17 (2005) R705-R774.

[12] R.B. Fair, Digital microfluidics: is a true lab-on-a-chip possible, Microfluid Nanofluid, 3 (2007) 245-281.

[13] T.B. Jones, On the relationship of dielectrophoresis and electrowetting, Langmuir, 18 (2002) 4437-4443.

[14] T.B. Jones, K.L. Wang, Frequency-dependent electromechanics of aqueous liquids: electrowetting and dielectrophoresis, Langmuir, 20 (2004) 2813-2818.

[15] K.L. Wang, T.B. Jones, Frequency-dependent bifurcation in electromechanical microfluidic structures, J Micromech Microeng, 14 (2004) 761–768.

[16] T.B. Jones, M. Gunji, M. Washizu, M.J. Feldman, Dielectrophoretic liquid actuation and nanodroplet formation, J Appl Phys, 89 (2001) 1441-1448.

[17] F. Mugele, Fundamental challenges in electrowetting: from equilibrium shapes to contact angle saturation and drop dynamics, Soft matter, 5 (2009) 3377-3384.

[18] W.Q. Wang, T.B. Jones, D.R. Harding, On-chip double emulsion droplet assembly using electrowetting-on-dielectric and dielectrophoresis, Fusion Sci and Technol, 59 (2011) 240-249.

[19] W.Q. Wang, T.B. Jones, Microfluidic actuation of insulating liquid droplets in a parallel-plate device, J Phys: Conf Ser, 301 (2011) 012057.

[20] J. Berthier, Ph. Clementz, J.M. Roux, Y. Fouillet, C. Peponnet, Modeling microdrop motion between covered and open regions of EWOD microsystems, Proc 2006 Nanotech Conf, (2006) 685-688.

[21] G. Wang, D. Teng, S.K. Fan, Three-dimensional digital microfluidics and applications, Proc IEEE NEMS, (2012) 415-418.

[22] A. Tuteja, W. Choi, M. Ma, J.M. Mabry, S.A. Mazzella, G.C. Rutledge, G.H. McKinley, R.E. Cohen, Designing superoleophobic surfaces, Science, 318 (2007) 1618-1622.

[23] T. Wu, Y. Suzuki, Engineering superlyophobic surfaces as the microfluidic platform for droplet manipulation, Lab Chip, 11 (2011) 3121-3129.