Microfluidic actuation of insulating liquid droplets in a parallel-plate device

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Abstract. In droplet-based microfluidics, the simultaneous movement and manipulation of dielectric and aqueous droplets on a single platform is important. The actuation forces on both dielectric and aqueous droplets can be calculated with an electromechanical model using an equivalent RC circuit. This model predicts that dielectric droplet actuation can be made compatible with electrowetting-based water droplet manipulation if the oil droplet is immersed in water. Operations such as transporting, splitting, merging, and dispensing of dielectric droplets at voltages less than 100 V are demonstrated in a parallel-plate structure. Such capability opens the way to fully automated assembly line formation of single-emulsion droplets.

1. Introduction
Among methods to move and manipulate liquid microdroplets, electrical actuation has many advantages, principally voltage-driven process control [1]. For the parallel-plate configuration shown in Fig. 1, electric actuation is attributed to electrowetting (EWOD) for conductive droplets and dielectrophoresis (DEP) for dielectric droplets. Both mechanisms drive droplets toward an activated electrode. There is a frequency-dependent relationship between EWOD and DEP [2]. For a liquid with finite electrical conductivity and dielectric constant, a critical frequency separates conductive (EWOD) and dielectric (DEP) limits [2-4].

Integration of EWOD and DEP-based droplet microfluidics is an essential step to forming double-emulsion droplets on a chip [5,6]. Unfortunately, DEP usually requires much higher voltage than EWOD. Low-frequency AC in the range of 11 to 100 V is sufficient for EWOD [7-9], but ~200 V typically is needed for DEP microfluidic actuation [3,5]. Here, we propose a low-voltage droplet manipulation scheme that uses the negative DEP effect to move dielectric droplets. A general

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electromechanical model predicts that dielectric droplet actuation can be made compatible with electrowetting-mediated water droplet manipulation. Companion experiments demonstrate the electric actuation of dielectric droplets, including droplet transport, splitting, merging, and dispensing at less than 100 V.

2. Modeling

Fig. 1 shows the cross-section of a typical microfluidic system consisting of two parallel glass plates. This structure is modeled by the equivalent RC circuits shown in Fig. 2. This RC circuit is similar to those of Jones [2-4, 10], Zeng [11], and Chatterjee [12]. Previous models describe the ambient medium with pure capacitance because the medium is usually air or oil. Here, however, the medium might also be conductive, so resistance is needed in parallel with the capacitance. The electromechanical force parallel to the electrodes is evaluated using coenergy \( W_e = C(x)V^2/2 \) [13].

Fig. 3 plots calculated forces for three cases: water droplet in air, mineral oil droplet in water, and mineral oil droplet in air, all at 65 V with substrate spacing of 170 \( \mu \)m. The electric field distribution in the liquid depends on frequency. In fact, for the water-in-air and oil-in-water cases there exists a critical value \( f_c \) that defines the transition from effectively conductive to dielectric behavior. See Fig. 3. There is no frequency dependence for the oil-in-air case because neither is electrically conductive.

The force acting on an oil droplet in air is much smaller than the force experienced by a water droplet at the same applied voltage. It is for this reason that oil droplet actuation usually requires higher voltages. On the other hand, an oil droplet in water medium experiences a relatively large electric force, comparable in fact to a water droplet in air. Note, however, that this force acts in the opposite direction, that is, from high to low field intensity. This is an example of negative DEP. The liquid droplet has a dielectric constant smaller than that of the surrounding medium. Though this force acts in the opposite direction, we can still achieve low voltage oil actuation by effectively pushing the droplet away from activated electrodes. Continuous movement becomes achievable by sequential activation and de-activation of electrodes in the array.

3. Fabrication and experimental setup

The basic features of the droplet-based microfluidic system are illustrated in Fig. 1. The bottom glass substrate is evaporatively coated with 100 nm aluminum, and then photolithographically patterned into a two dimensional array of individually addressable electrodes. The plate is then spin-coated with 0.5 \( \mu \)m of spin-on-glass (SOG) (Futurrex IC1-200) as the dielectric layer and 1 \( \mu \)m of amorphous fluoropolymer (DuPont Teflon-AF) as a hydrophobic coating. The upper plate is glass coated with transparent ITO (Indium-tin oxide), which forms the ground electrode, and top-coated with 1 \( \mu \)m Teflon-AF as a hydrophobic coating. The top substrate is positioned above the bottom substrate using spacers. Voltages applied to individual electrodes are controlled by a LabView-based controller.
4. Results and discussion

The layout of the basic design of a transport array for oil droplets in water is illustrated in Fig. 4. The side electrodes help contain the droplets on the track. To perform an experiment, an aqueous solution is first introduced into the gap between the top and bottom substrates. The surfactant Silwet L-77 (0.0625 wt%) is used in the water phase to reduce surface tension and stabilize the mineral oil droplet in the water medium. The oil droplet is then injected.

**Oil droplet transport**  Fig. 5 shows video frames demonstrating operation of the oil droplet transport scheme in water. The droplet initially sits on electrode 1, as shown in Fig. 5 (a). All the electrodes are initially off. When electrode 1 and the side electrodes are activated, the oil drop is pushed to the left. See Fig. 5 (b). After the droplet reaches electrode 2, this electrode is now activated forcing the oil droplet to continue moving left to the next electrode. In this way, continuous movement is easily achieved.

**Oil droplet splitting and merging**  After shown in Fig. 6, droplet division is achieved by activating the electrode on which the droplet resides while deactivating its two neighbors. In Fig. 6 (a), a large oil droplet is initially sitting on electrode 2 as shown. By activating electrode 2, the oil drop starts to move outwards onto electrode 1 and 3. See Fig. 6 (b). Eventually the oil drop splits in two. Merging is achieved simply by moving two oil droplets together. When electrodes 1 and 3 are activated, the two small oil drops are pushed back together onto electrode 2 where they quickly coalesce.

**Oil droplet dispensing**  Fig. 7 provides a demonstration of oil droplet dispensing from a large reservoir. The oil initially covers the large reservoir electrode. When this electrode is activated, with the side electrodes also turned on, oil is drawn outward onto the transport array to form a liquid column. See Fig. 7 (b). Liquid pinch-off is achieved by activating the electrode next to the reservoir and deactivating the reservoir electrode. See Fig. 7 (c). The portion of the liquid to the left pinches off into a droplet which can then be transported away. The process may be repeated to dispense a stream.
of droplets. Note that sometimes a very thin film of oil connecting the dispensed droplet to the reservoir forms for a time. This film eventually ruptures.

5. Conclusions

The general electromechanical model developed here can be used to calculate electric actuation forces on both dielectric (that is, insulating) and aqueous (conductive) droplets in parallel-plate microfluidic structures. The model explains why oil droplet actuation requires higher voltage than does an aqueous droplet. It also confirms the possibility of low voltage, negative DEP actuation of oil when the droplet is in a water medium. These modeling predictions are corroborated by experiments that demonstrate some essential droplet-based microfluidic operations, including oil droplet transport, splitting, merging, and dispensing. These operations can be monolithically integrated on a single microfluidic chip to provide a versatile platform for the simultaneous processing of many different types of liquids at relatively low voltages. One particular application of interest for the assembly-line fabrication of the targets for laser fusion experiments is an automated double-emulsion droplet generator.

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