Microfluidic electrowetting-based droplet mixing

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Abstract- We present a liquid droplet mixer to enable mixing of the samples and reagents for chemical and biological analysis in micro total analysis systems (µTAS). The droplets in the mixer are actuated based on electrowetting phenomenon. The actuator comprises of two parallel glass plates between which the droplet is actuated on a planar array of electrodes. Mixing of liquid channels in microfluidic systems has been demonstrated for continuous flow systems where the mixing is diffusion-limited due to the laminar flow of liquids, requiring very long, thin channels.

In the present paper, mixing is performed on discrete droplets of liquid. When two droplets are brought together, depending on the velocity of the moving droplets, surface tension, viscosity, electrode activation, and volume among other factors, turbulence is created which aids in mixing. The mixing is not limited by diffusion and enhanced by transport. The mixing experiments are performed between fluorescein and plain water droplets whose individual volume is 1.75µl. Mixing is visualized with a 2-CCD camera setup to observe both the top and side views, with appropriate filters to capture fluorescence. We observed that it takes about 60 seconds for two droplets of 1.75µl each to mix when their surface tensions are different and it takes about 90 seconds when the two droplets have similar surface tensions. The present mixer stands apart from any current conventional micromixers in that the mixing times occupies much lesser area, mixing does not need any specific architecture on the chip and can be performed on any transport electrodes dynamically assigned to mixing.

Index Terms— Droplet, electrowetting, fluorescence, microfluidics. mixing

I. INTRODUCTION

MIXING .in microfluidic devices is one of the critical steps in realizing a μ TAS (micro total analysis system) or for "lab on a chip" systems. Mixing requirements can be either in dilution of the samples before analysis or in mixing the samples with reagents in a particular ratio. The ability to mix liquids rapidly and utilizing minimum area, greatly improves the throughput of such systems.

We present a microfabricated electrowetting actuator that can be used to perform mixing, transport, and dispensing. Electrowetting is the process by which the interfacial tension between a liquid and solid phase is modulated electrically [1]. This device has all the advantages of a microfluidic system such as the ability to handle small volumes, high throughput, rapid transport, and batch fabrication. However, liquid is handled as droplets enabling discrete manipulation unlike conventional continuous flow microfluidic devices. Since the liquid is handled as droplets, just the right volume required for a reaction can be utilized thereby reducing the dead volume. In this paper, we present some preliminary results obtained on mixing in this device.

Mixing in microfluidics is performed either by turbulence or through interdiffusion. Since the Reynolds numbers are very low for continuous flow systems, mixing relies mainly on the interdiffusion in the channels. Several researchers have tried various approaches to create turbulence in the flow streams to promote mixing. Hosokawa et al. have demonstrated mixing of droplets in a hydrophobic microcapillary valve device where the droplets are formed, actuated, and mixed with the help of air pressure [2]. They mixed two droplets of fluorescein and deduced mixing from the fluorescence intensity measurements obtained from top and side view to get a more complete picture of the mixing process as it occurs. In our device, due to the nature of the motion of the droplet, which is not clearly understood yet, mixing is enhanced.

II. EXPERIMENTAL

The droplets are actuated by electrowetting phenomenon in which their surface tension is controlled electrically as demonstrated by Pollack et al [3]. A droplet of polarizable and conductive liquid is sandwiched between two planar electrodes. The bottom plate consists of an array of independently addressable chrome control electrodes coated with Parylene C (800 nm) for insulation while the top plate coated with Indium Tin Oxide (ITO) acts as a ground electrode. Both the electrode surfaces are coated with a thin hydrophobic layer of Teflon AF 1600 (50 nm). The top and bottom plates were separated by a glass spacer yielding a fixed gap.

Each droplet is made to contact both the top and bottom

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electrodes. The volume of the droplet is chosen to overlap the two adjacent bottom electrodes slightly. The sides of the bottom electrodes are interdigitated to increase the overlap of the droplet. The droplet is surrounded by either just ambient air or silicone oil, which is immiscible with the droplet. A custom control system was made to address and switch each electrode. Since this actuation does not need any fixed channels or moving parts, any electrodes on the chip can be designated for mixing.

The schematic of the droplet actuation for mixing is shown in top and side view in Fig.1. A droplet containing fluorescein is actuated towards a non-fluorescein droplet. The fluorescein droplet contains 1mM fluorescein (obtained from JT Baker) for fluorescence, 0.125M KCl for making the droplet conductive, and 0.125M NaOH since the fluorescence of fluorescein is pH — dependent. The non-fluorescein droplets contain 0.125M KCl and 0.125M NaOH or just 0.125 M KCl. All the droplets were made in deionized water.

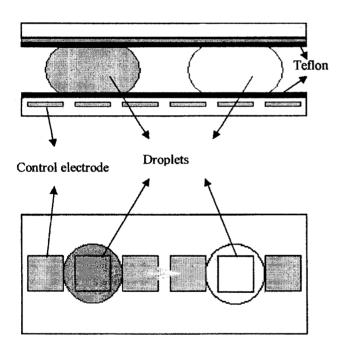


Figure 1 Schematic side and top view of the electrowetting-based mixing actuator

Either the fluorescein droplet is moved towards the non-fluorescein droplet or vice versa. The resulting motion is captured on a video through a CCD camera. A two-camera setup was used to get both the top and side view of the mixing process simultaneously as shown in Fig. 2. The fluorescein droplet was excited with a tungsten lamp with a blue filter (490 nm). Both the cameras were mounted with long pass filters (>510 nm) to collect the fluorescence. The videos were then digitized for further analysis.

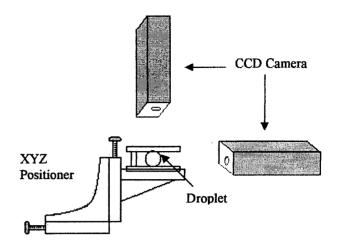


Figure 2 Two CCD camera setup to view top and side view of droplet mixing

The interfacial tension of a fluorescein droplet, containing 0.1mM fluorescein and 0.125M NaOH and KCl, with 1cSt silicone oil is measured to be 37 dynes/cm whereas a non-fluorescein droplet, containing only NaOH and KCl both at 0.125M, has 36 dynes/cm with the same oil. The viscosity for fluorescein and non-fluorescein droplets is 1.396 and 1.373 cP and the conductivity is 29.7 mMho & 25.9 mMho, respectively. The interfacial tension of a 0.125M KCl droplet is 31.4 dynes/cm with 1 cSt silicone oil.

III. RESULTS AND DISCUSSION

A number of parameters can be varied to study their effect on mixing in our current setup. For the initial experiments, we have chosen to fix the gap between the top and the bottom electrodes (i.e., the droplet height) at 800 µm and the pitch of the bottom electrodes as 1.5 mm, which fixes the volume of the droplet. The volume of each droplet was fixed at 1.75 µl in all the experiments. The voltage of actuation for the moving droplet also is fixed at 30V since the voltage affects the speed of the impinging droplet and thereby the mixing. All the experiments were performed with the surrounding medium of 1 cSt silicone oil.

Initially a fluorescein droplet containing fluorescein, NaOH and KCl (designated as F) was moved onto a non-fluorescein droplet containing NaOH and KCl (NF) with the appropriate activation of the sequence of electrodes as shown in Fig.1. The electrode adjacent to NF droplet is switched on and off for a very small amount of time for the F droplet to move onto the NF droplet so that both the F and NF droplet finally occupy the same electrode which NF was initially occupying. In this case, there is a very small difference in the interfacial tension of the droplets with oil. Even though the droplets look

mixed from the top view in about 15 seconds, the view from the side reveals that F droplet has just gone underneath the NF droplet as shown in Fig. 3. Mixing proceeds through diffusion from this point and it takes about 90 seconds to completely mix.

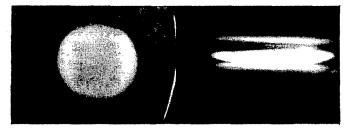


Figure 3 Top and Side views of a fluorescein droplet mixing with a non-fluorescein droplet at 15 seconds after coalescence.

If the initial coalescence does not create enough turbulence for mixing then we observe that the F droplet and NF droplet are vertically separated. At this point, the mixing is just diffusion limited. To enhance the mixing rate, the area of contact was increased. Experiments were performed similar to the previous one but the difference being the coalescence occurs on two electrodes instead of one electrode. The F and NF droplet come together on two electrodes which doubles the area of contact compared to the previous case. We do not observe a significant change in the mixing times in this case.

In the next set of experiments, we moved an F droplet towards a KCl droplet (which has just 0.125M KCl). It was repeatedly observed that the F droplet would just engulf the KCl droplet while the droplets are coalescing. From the side view, it would appear as if mixing is complete since the KCl droplet is completely engulfed by the F droplet. However, from the top view we observed that the F droplet forms a ring around the KCl droplet yielding a donut shape as shown in Fig 4. Within 10 seconds, uniform fluorescence can be observed from the top but from the side view it appears that fluorescein goes underneath the KCl droplet as shown in Fig. 5. From now on, mixing seems to occur by diffusion. Uniform fluorescence appears from the side in 1 second, from the top in 10 seconds, and again from the side in 60 seconds from the start of coalescence.



Figure 4 Top and side views of a fluorescein and KCl droplet immediately after coalescence.



Figure 5 Top and side views of a fluorescein and KCl droplet 10 seconds after coalescence

A similar experiment was performed with the KCl droplet moving into the Fluorescein droplet. In this case, the droplets just come together and there is no engulfinent as in Fig. 4. Nevertheless, it takes just about the same time for mixing to finish. It appears that mixing is faster in droplets, whose surface tensions are very different. When the surface tensions are similar then it takes about 90 seconds (1.5 times more than surface tension mismatched case) for mixing to finish and when the surface tensions are similar and the coalesced droplet is spread on two electrodes then it takes 180 seconds (3 times more than surface tension mismatched case) for mixing to finish.

It should be noted that we are observing only from two angles, which may not yield sufficient information. Ideally, we would like to get a 3D view of the droplet while it is mixing to see if there are any unmixed volumes in the coalesced droplet, which cannot be made out by just looking at the surface of the droplets from all angles.

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