COALESCENCE AND MIXING OF DROPLETS WITH IDENTICAL AND DISTINCT SURFACE TENSIONS ON A WETTABILITY GRADIENT SURFACE

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ABSTRACT
This study investigates the influence of identical and distinct surface tensions on droplet coalescence and mixing processes after a head-on collision between a moving droplet and a stationary droplet on a wettability gradient surface (made from the self-assembly monolayer, SAM technique) by micro-PIV (particle image velocimetry) measurement and micro-LIF (laser induced fluorescence) technique. The results demonstrate that coalescence of the droplets with distinct surface tensions achieves superior mixing quality to that of the droplet with identical surface tension. This study would significantly contribute to the issue of droplet mixing and reaction in a microfluidic open system.

KEYWORDS: Wettability gradient surface, Droplet coalescence, Mixing, Micro-PIV

INTRODUCTION
Droplet-based microfluidic system has drawn much attention for its potential applications in optics, electrics, and micro total analysis systems (μ-TAS). It is expected to revolutionize biological laboratory procedures by allowing faster and more error-free assays, where droplets are biological sample carriers [1]. Transport, collision, and mixing of droplets are the key issues for the development of droplet-based microfluidic systems.

Mixing is an imperative mechanism that crucially impacts biochemical reactions in digital-microfluidic systems [2], and attracts much research attention. The majority of droplet-based mixers are so far either consisted of microchannels or of electrowetting devices which not belong to an open system; the studies about the phenomena of droplets mixing on an open microfluidic system are less addressed. In addition, the fluid with different surface tensions have wide range for biochemical reagent might be the main factors influence droplet collision and mixing process. The issues about how surface tension affect droplet collision and mixing behavior are also less addressed. In this study, the droplets with different and distinct surface tensions were used in an open droplet manipulating system. We observed the spontaneously transport and droplet collision process on a wettability gradient surface. Micro particle image velocimetry (micro-PIV) and confocal fluorescence microscopy (CFM) were employed to analyze the effect of surface tension on coalescence and mixing of two droplets on a wettability gradient surface manufactured using the self-assembly monolayer (SAM) technique [3,4].

EXPERIMENTAL
The modified vapor diffusion self-assembled monolayer (SAM) method was used to fabricate the wettability gradient surface on a glass plain microscope slide so that light source of micro-PIV and micro-LIF and their signals can go through from the bottom. The experimental apparatus is also sketched in Figure 1. Droplets which were generated by a syringe pump (KDS220, KD Scientific) with microliter syringes (Hamilton 710RN) had volume of 300±12 nL. One stationary droplet was positioned in the hydrophilic region of the surface, and a moving droplet initially situated in the hydrophobic region was transported to collide with the stationary droplet.

A high-speed camera (IDT X-stream) with the objective lens captured the views at a rate of 1000 fps with a resolution of 2352 x 1782 pixels (2.1 x 1.6 mm) to be micro-PIV measurement and calculation. Micro-PIV is used to investigate the internal flow induced from the coalescence of droplet. The images captured by camera were process with a standard cross-correlation scheme (Insight 5, TSI Inc.) to produce the raw vector field. This scheme implemented a multi-pass interrogation in which in the first pass cross-correlation was calculated for an interrogation spot of area 64 x 64 pixel; the interrogation spot was then divided into four subareas 32 x 32 pixel for the second pass calculation. The interrogation spots overlapped by half; a Gaussian fit was used to resolve the sub-pixel displacement.

The mixing efficiency and mixing pattern inside the coalesced droplet was obtained by micro-LIF technique. The fluorescence DNA solutions of FAM-labeled oligonucleotide (6-carboxy-fluorescein, 2 μM, 495/521 nm) and Cy-5-labeled oligonucleotide (cyanine dye, 2 μM, 646/662 nm) were adopted for simulating the mixing of biochemical fluids. The excitation lights, provided from an argon laser (488 nm) and a diode laser (638nm), were directed into an objective lens (Plane Apochromat VC 10 x, NA = 0.45) with a dichroic mirror, by which the excitation lights and emitted lights were separated. A pinhole (diameter 12.8 μm) was installed behind the dichroic mirror to optically cut off light from out-of-focus plane. The intensities of the emitted lights were measured with two separate photomultiplier tubes (PMT) behind the filter blocks. Using the xyz stage
to scan several z-positions, the 3D reconstructed distribution of fluids can be pictured due to the high vertical resolution of confocal microscopy.

In this work, working fluids of three types were utilized for four different experimental cases (Table 1). To produce fluids with various surface tensions, Tween 20® (polyoxyethylene sorbitan monolaurate, Pierce Chemical Co.), a kind of surfactant, which can reduces surface tension in a fluid were added into DI water. Surface tensions were measured using an EZ-Pi Surface Tensiometer (Kibron Inc. Helsinki, Finland). Four different cases performed in this study are also listed in Table 1.

Table 1. Surface tensions and contact angles on the hydrophilic region (glass substrate) and the hydrophobic region (a hydrophobic SAM layer, DTS, on the substrate) of two working fluids.

<table>
<thead>
<tr>
<th>Working Fluids</th>
<th>surface tension (mN/m)</th>
<th>at hydrophobic region θ (degree)</th>
<th>at hydrophilic region θ (degree)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Working fluid A</td>
<td>73.28</td>
<td>106.7</td>
<td>59.2</td>
</tr>
<tr>
<td>Working fluid B</td>
<td>46.51</td>
<td>72.5</td>
<td>26.7</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Experimental Cases</th>
<th>stationary droplet</th>
<th>moving droplet</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case A</td>
<td>working fluid A</td>
<td>working fluid A</td>
</tr>
<tr>
<td>Case B</td>
<td>working fluid B</td>
<td>working fluid B</td>
</tr>
<tr>
<td>Case C</td>
<td>working fluid A</td>
<td>working fluid B</td>
</tr>
<tr>
<td>Case D</td>
<td>working fluid B</td>
<td>working fluid A</td>
</tr>
</tbody>
</table>

RESULTS AND DISCUSSION

Figure 2 manifests the dominant flow patterns during the coalescence of droplets. For the case A, a right-to-left central flow was observed 1 ms after the droplet collision was initiated; a pair of symmetric vortices was subsequently formed 5 ms later and till to 15 ms. The flow patterns for the case B were analogous to that for the case A; however, the vortex strength was comparatively weak due to the smaller surface tension of the fluids. For the case C, horizontal vibration of the flow field inside the coalescing droplet occur immediately after the collision. Since the surface tension (cohesion) of the stationary droplet was greater than that of the moving droplet, the formation of vortices in the coalescing droplet was suppressed. For the case D, as the moving droplet possessed a relatively large cohesion, the induced vortices were kept at the right side of the coalescing droplet. The duration of vortices was estimated small due to the relatively small surface free energy.

Mixing patterns and transient mixing indices of the coalesced droplet for the cases A – D are respectively shown in Figure. 3. Each data point in Figure. 3(b) represents the averaged mixing index over the 3D scanning duration. The complicated mixing pattern of the case A was mushroom-shaped (constituted by recirculation flows). For the case B, a portion of the moving droplet was unmerged with the stationary droplet because of the relatively low surface tension of the droplets; the efficacy of the droplet mixing was therefore poor, rendering relatively small mixing indices. For the case C, the mixing pattern illustrates that the fluid of the moving droplet was surrounded by the fluid of the stationary droplet through the cohesion of the stationary droplet acting on the moving droplet; the trend of the mixing indices were equal to that for the case A. The mixing pat-
terns for the case D exhibits that the fluid of the stationary droplet was separated by that of the moving droplet through the effect of the cohesion of the moving droplet on the stationary droplet; consequently, the interfacial area of the fluids was extended, thus promoting the mixing of the fluids. The slope of the mixing indices for the case D is relatively sharp, demonstrating that the extended interface enhanced the diffusion for fluid mixing.

Figure 2. The representative patterns of internal flow during the droplet coalescence for cases A~D.

CONCLUSION

We demonstrated that surface tensions of droplets has substantial impacts on their coalescence and mixing; the droplets with distinct surface tensions result in a better mixing quality than that with identical surface tension during coalescing. The results of this work would serve as a significant reference for the digital microfluidic system.

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REFERENCES


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