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Droplet Pattern Formation and Translation in New Microfluidic Flow-Focusing Devices

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We conducted experiments on specially designed microfluidic chips that generate droplets through a microfluidic flow-focusing approach. The fluid flow in the microfluidic channel produced a shear flow field at low Reynolds numbers. The droplets in the microfluidic system exhibited special droplet pattern formations similar to periodic crystal-like lattices because of the competition between shear forces and surface tension. By adjusting the flow rate ratio of the water (droplet phase) to oil (continuous phase) phases and changing the outlet channel widths, the droplets formed monolayer dispersion to double-layer formation to monolayer squeezing when the outlet channel widths were 250 or 300 µm. We also obtained droplets with monolayer dispersion, three-layer arrangements, double-layer squeezing, and monolayer squeezing when the outlet channel width was 350 µm. The outlet channel width was increased to 400 µm, and four-layer arrangements were observed. We also studied the translation of droplet formation, which resulted in a detailed strategy to control drop size and droplet pattern formation for emulsification in microfluidic devices. We expect that our strategy can provide theoretical guidance to synthesize dispersion or polydisperse colloid particles.

Key words: Microfluidic flow-focusing device, Droplet pattern formation, Transition

I. INTRODUCTION

Microfluidics is a novel technology that deals with precise control and manipulation of small-volume fluids in channels with submillimeter scales. Extensive efforts have been devoted to applying biotechnological and chemical reactions in microfluidic devices, such as polymerase chain reaction \cite{1, 2}, DNA analysis and sequencing \cite{3, 4}, protein crystallization \cite{5, 6}, cell culture and analysis \cite{7–9}, and biomedical diagnosis \cite{10, 11}. Droplets generated through microfluidic emulsification have more advantages than those created through traditional emulsion because the droplet size and size distribution of the former can be controlled at a micro scale.

Therefore, these droplets have been used in a wide range of applications and have been extensively studied because of their various unique advantages. These droplets have been actively investigated from high-throughput droplet PCR \cite{12, 13} to cell encapsulation \cite{14}, colloidal particle production \cite{15–17}, and droplet formation \cite{18–21}.

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Investigations during the past two decades have mainly focused on preparing monodisperse and polydisperse droplets with precisely controlled sizes and distributions by using different microfluidic devices, such as T-junction and flow-focusing microfluidic devices. Investigations on droplet formation are also important for emulsification. Bubble or droplet formation in microfluidic devices, bubble or droplet formation in T-junction or flow-focusing microfluidic devices have been investigated \cite{22–24}. New microfluidic generators were invented to achieve O/W/O double emulsions or form water droplets with high-volume fractions of gas bubbles \cite{25}. The significant difference in droplet sizes between using flow-rate-controlled (syringe pumping) and pressure-controlled flows has been studied \cite{26}. Droplet size changes only slightly, and droplet speed increases linearly, with the flow rate ratio of the water (droplet phase) $Q_1$ in a flow rate-controlled flow. Dripping to jetting transitions have also been studied in co-flowing microfluidic \cite{27}.

We report new droplet pattern formations in the microfluidics devices (FFDs). The droplets form periodic crystal-like lattices. After adjusting the flow rate ratio of the water (droplet phase) and oil (continuous phase) phases $Q_1/Q_2$, the droplets form monolayer dispersion to double-layer formation to...
monolayer squeezing when the outlet channel is 250 or 300 μm. Moreover, we observe triple- and quadruple-layer formations when the outlet channel widths are 350 and 400 μm, respectively. The droplet formation dynamic process is described analytically.

II. EXPERIMENTS

Poly(dimethylsiloxane) elastomer (Sylgard 184) was purchased from Dow Corning. Light mineral oil and sorbitan monoooleate (SPAN-80) were supplied by Sigma-Aldrich. Negative photoresist SU-8 2050 was purchased from MicroChem. Silicon wafers were supplied by New Semiconductor Manufacturing Co., Ltd.

A. Microfluidic device fabrication

Microfluidic experiments were carried out using standard soft photolithography techniques [28, 29]. A negative photoresist SU-8 2050 was used to fabricate the channel mold on silicon wafers. The channels were made of PDMS, which was poured over the channel mold and left to harden at 85 °C for 1 h. The channel was then treated with oxygen plasma and bonded to another thin bottom sheet of PDMS. Finally, a microfluidic chip was successfully fabricated.

B. Droplet generation in microfluidic FFDs

Microfluidic FFDs were used to generate droplets. Figure 1(a) shows that the cross section of the channels is rectangular, and the channel height is 50 μm. The width of the water inlet channel was \( W_i = 150 \) μm and that of the oil inlet was \( W_o = 100 \) μm. The orifice was \( D = 60 \) μm. Four similar microfluidic FFDs (FFD-1, FFD-2, FFD-3, and FFD-4) were integrated in one microfluidic chip with outlet channel widths of 250, 300, 350, and 400 μm. The liquids were forced through a narrow orifice in which a thread of water broke up and released the droplets [30, 31]. Droplet-phase deionized water and two streams of continuous-phase mineral oil (Sigma-Aldrich) were injected into the central and side channels, respectively. The water broke up, and the droplets were generated because of the flow shear stress.

C. Emulsification experiments in microfluidic FFDs

Liquids were injected into the microfluidic devices by using polyethylene (Smiths Medical International, PE 0.86 mm ×0.205 mm ). The tubes were connected from the syringe needle to the inlet hole of the device channel by three syringe pumps (syringe pumper, NE-1000). A CCD camera (Photometrics CoolSNAP ES) coupled with an inverted microscope (Olympus IX71) was used to capture images of the droplets. The size and distance of the droplets were determined through image analysis by using the software PixeLINK uscope. The emulsification dynamic process was also observed directly using a video camera with an image capture software (PixeLINK Capture OEM).

III. RESULTS AND DISCUSSION

The microfluidic chip integrated FFD-1, FFD-2, FFD-3, and FFD-4, which exhibited higher efficiency and stability for generating droplets than the T-junction microfluidic system. The emulsification process was studied by changing \( Q_i/Q_o \) of the water and oil phases. The water driven by the syringe pump was forced through a narrow orifice by two streams of oil in the side channel during the interaction of shear stress and surface tension. The water became a thread and was then broken up to release droplets. The micron-sized droplets in the microfluidic system exhibited a special phenomenon. The fluid (liquid) displayed and produced a macro scale of different special phenomena because of micron-grade structure. Two emulsification processes were studied by adjusting \( Q_i/Q_o \) and changing the outlet channel widths. The oil phase flow rate was uniformly set to 0.1 mL/h. The flow rate of the water phase was increased by 0.01 mL/h. We observed droplets that formed monolayer dispersion to multilayer arrangements to monolayer squeezing in the out-

FIG. 1 Schematic of microfluidic FFDs used to generate droplets. (a) The width of the water inlet channel was \( W_i = 150 \) μm, the oil inlet was \( W_o = 100 \) μm, and the orifice was \( D = 60 \) μm. The flow rate of the water phase is \( Q_i \), while the flow rate of the oil phase is \( Q_o \). The outlet channel widths is \( W_o \). (b) Images of microfluidic FFDs (top view). The four widths of the outlet channels were A: FFD-1, 250 μm; B: FFD-2, 300 μm; C: FFD-3, 350 μm and D: FFD-4, 400 μm. The channel height was 50 μm.
let channel by changing $Q_i/Q_o$ when the outlet channel widths $W_i$ were 250 and 300 $\mu$m. We obtained droplets with three- and four-layer arrangements when the outlet channel widths $W_i$ were 350 and 400 $\mu$m, respectively.

A. Changing $Q_i/Q_o$ when $W_i$ are 250 and 300 $\mu$m

FFD-1 and FFD-2 were used in the experiment, with outlet channel widths $W_i$ of 250 and 300 $\mu$m, respectively. Changing $Q_i/Q_o$ resulted in similar droplet pattern formations in both FFDs. We analyzed FFD-1 in detail.

Figure 2 shows that the droplets have three shapes: spherical, discoid, and long-striped. We endeavored to identify the volume of these different droplets. The volume $V$ of these three droplet shapes was different. Figure 2(a) shows that the first droplet shape was spherical and undeformed. Volume $V_1$ is easily given by

$$V_1 = \frac{3}{4} \pi \left( \frac{d}{2} \right)^3$$

where $d$ is the droplet diameter. The second droplet shape was a discoid. Figure 2(b) shows that the diameter of an undeformed droplet $K$ was greater than the channel height $h$. The volume of the discoid droplets $V_2$ was approximated as

$$V_2 = \frac{\pi}{12} [2K^3 - (K - h)^2(2K + h)]$$

When $Q_i/Q_o$ was increased, long-striped droplets with semicircular ends appeared. Figure 2(c) shows volume $V_3$ with $M$ and $N$ as the length and width of the long-striped droplets, respectively. The droplet volume was calculated as

$$V_3 = \left[ \pi \left( \frac{N}{2} \right)^2 + MN \right] h$$

Water was forced though the orifice in the microfluidic FFDs, which then broke up and released droplets. As soon as the droplets formed, their velocity of water was higher than that of the oil. The water velocity then decreased as the droplets flowed away from the orifice and became equal to that of the oil. Thus, when the droplets were far away from the orifice within time $t$, their velocity was as follows:

$$v_1 = \frac{Q_i + 2Q_o}{W_0 h} = v_o$$

where $v_1$ is the droplet velocity, and $v_o$ is the velocity of oil.

The volume of droplets $V_{1,2}$ is expressed as the volume of a spherical undeformed droplet or discoid droplet. Therefore, within time $t$, we easily obtain the number of droplets $n$

$$n = \frac{Q_i t}{V_{1,2}}$$

Droplets flow through path $l$ within time $t$, which is easily deduced as

$$l = tv_i$$

The distance $f$ along the flow direction between the edges of two adjacent droplets can be given by

$$f = \frac{l - dn}{n - m}$$

where $m$ is the number of droplet layers. As seen in Fig.3, the schematic shows that the parameters ($f$, $m$, $n$, $d$, and $l$) are directly associated with the droplet patterns of monolayer and multilayer dispersion. $f$ is different from monolayer layer dispersion to multiple layers. When droplets appeared monolayer layer dispersion ($m=1$), $f$ is expressed as $f=\frac{l - dn}{n - 1}$. When droplets exhibited multiple layer dispersion ($m=s\geq2$), $f$ is shown as $f=\frac{l - dn}{n - s}$.

In microfluidic FFDs, the droplets exhibited special droplet pattern formations because of competition between shear forces and surface tension. A significant
kinetics constant capillary number \((C_a)\) was generally introduced:

\[
C_a = \frac{\mu v_o}{\sigma}
\]

where \(\mu\) is the rate of shear forces and surface tension, \(v_o\) is the velocity of the oil phase, and \(\sigma\) is the surface tension of the water and oil phases. The shear forces and surface tension dominated two regimes. When \(Q_1/Q_o\) was small, \(C_a\) was small and the surface tension dominated. The droplets appeared as multilayer dispersions and exhibited spherical droplets or discoid droplets to minimize the droplet surface area. When \(Q_1/Q_o\) was large, \(C_a\) was large and the shear force dominated. The droplets appeared as monolayer squeezing and formed long-striped droplets. Meanwhile, two transitions of the droplet pattern formation occurred.

Surface tension dominated when \(Q_1/Q_o\) was small. Droplets displayed monolayer dispersion and double-layer arrangements without squeezing between droplets. Figure 4(a) shows that the droplet shape was spherical by undeformed when \(0<Q_1/Q_o<0.3\). Moreover, the droplets appeared in a monolayer dispersion arrangement without squeezing between droplets, and one droplet layer \((n=1)\) existed. Thus, \(f=n-1\).

When \(Q_1/Q_o\) increased, \(f\) progressively decreased until it equaled \(0\) \((Q_1/Q_o=0.3)\). Figure 4(b) shows that the first droplet pattern formation transition appeared when \(0.3\leq Q_1/Q_o<3.5\). The droplet shape was discoid, and the droplets displayed a double-layer droplet arrangement \((n=2)\). Thus, \(f=\frac{l-dn}{n-2}\). No obvious squeezing phenomenon was observed.

Figure 4(c) shows that the shear forces on the droplet were sufficiently large to overcome the surface tension in the second regime when the flow rate ratio was \(Q_1/Q_o\geq3.5\). Droplets formed a monolayer squeezing arrangement. Meanwhile, the second significant droplet pattern formation transition occurred. Droplet morphology was markedly different, which resulted in long-striped droplets because of the squeeze between droplets.

The droplet pattern formation in the outlet channels of FFD-1 with \(W_4=250\ \mu m\) was markedly different with increasing of \(Q_1/Q_o\). Two significant droplet formation transitions occurred. First, the droplets appeared in a monolayer dispersion arrangement when the \(Q_1/Q_o\) was small. Second, increasing \(Q_1/Q_o\) resulted in a double arrangement of droplet pattern formation. The droplet pattern formation exhibited a monolayer squeezing arrangement when \(Q_1/Q_o\) was large enough. FFD-2, which had an outlet channel width \(W_4\) of \(300\ \mu m\), had a similar droplet pattern formation to FFD-1. When \(Q_1/Q_o\) of FFD-2 was increased, monolayer dispersion arrangement, double dispersion arrangement, and monolayer squeezing arrangement occurred.

\section{B. Changing \(Q_1/Q_o\) when \(W_4=350\ \mu m\)}

When \(W_4\) of FFD-3 was \(350\ \mu m\), droplet pattern formation changed by increasing the flow rate ratio of \(Q_1/Q_o\) as shown in Fig.5. The droplet pattern formation exhibited competition between shear forces and surface tension in the microchannels. shear forces and surface tension dominated two respective regimes. However, three transitions of droplet pattern formation occurred. One transition of droplet pattern formation occurred. When \(Q_1/Q_o\) was small, \(C_a\) was small and surface tension dominated. The droplet pattern formation then resulted in monolayer dispersion and three-layer dispersion arrangement without squeezing between droplets. Figure 5(a) shows that the droplet shape was spherical undeformed when \(0<Q_1/Q_o<0.1\), and droplets appeared in a monolayer dispersion arrangement. The distance \(f\) along the flow direction between the edges of adjacent droplets was given by \(f=\frac{l-dn}{n-1}\) \((n=1)\). When \(Q_1/Q_o\) was increased, \(f\) de...
TABLE I Different droplet pattern formations and droplet shape with different outlet channel widths for FFD-1, FFD-2, FFD-3, and FFD-4 by changing the flow rate ratio $Q_i/Q_o$.

<table>
<thead>
<tr>
<th>Device</th>
<th>$W_d/\mu m$</th>
<th>$Q_i/Q_o$</th>
<th>Droplet pattern</th>
<th>Droplet shape</th>
</tr>
</thead>
<tbody>
<tr>
<td>FFD-1</td>
<td>250</td>
<td>0&lt;(Q_i/Q_o)≤0.3</td>
<td>Monolayer dispersion</td>
<td>Spherical</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.5&lt;(Q_i/Q_o)≤3.5</td>
<td>Double-layer dispersion</td>
<td>Discoid</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.5&lt;(Q_i/Q_o)</td>
<td>Monolayer squeezing</td>
<td>Long-striped</td>
</tr>
<tr>
<td>FFD-2</td>
<td>300</td>
<td>0&lt;(Q_i/Q_o)≤0.5</td>
<td>Monolayer dispersion</td>
<td>Spherical</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1&lt;(Q_i/Q_o)≤3.6</td>
<td>Double-layer dispersion</td>
<td>Discoid</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.6&lt;(Q_i/Q_o)</td>
<td>Monolayer squeezing</td>
<td>Long-striped</td>
</tr>
<tr>
<td>FFD-3</td>
<td>350</td>
<td>0&lt;(Q_i/Q_o)≤0.1</td>
<td>Monolayer dispersion</td>
<td>Spherical</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1&lt;(Q_i/Q_o)≤3.5</td>
<td>Double-layer dispersion</td>
<td>Discoid</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.5&lt;(Q_i/Q_o)≤4</td>
<td>Double-layer squeezing</td>
<td>Long-striped</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4&lt;(Q_i/Q_o)</td>
<td>Monolayer squeezing</td>
<td>Long-striped</td>
</tr>
<tr>
<td>FFD-4</td>
<td>400</td>
<td>0&lt;(Q_i/Q_o)≤0.025</td>
<td>Monolayer dispersion</td>
<td>Spherical</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.1&lt;(Q_i/Q_o)≤0.5</td>
<td>Double-layer dispersion</td>
<td>Discoid</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2&lt;(Q_i/Q_o)</td>
<td>Four-layer dispersion</td>
<td>Discoid</td>
</tr>
</tbody>
</table>

Therefore, the droplet pattern formation was markedly different for those with $W_d=350 \mu m$ from those with $W_d=250$ and 300 $\mu m$ with increasing $Q_i/Q_o$. First, the droplets appeared in a monolayer dispersion arrangement when $Q_i/Q_o$ was small. Second, the droplets appeared in a three-layer dispersion arrangement when $Q_i/Q_o$ increased sufficiently. Third, the droplet pattern formation exhibited a double-layer squeezing arrangement when $Q_i/Q_o$ increased excessively. The final result was a monolayer squeezing arrangement.

C. Changing $Q_i/Q_o$ when $W_d=400 \mu m$

The $W_d$ of FFD-4 was 400 $\mu m$. Figure 6 shows that the droplet pattern formation changed with increasing $Q_i/Q_o$. Figure 6(a) shows that the droplets appeared in a monolayer dispersion arrangement when $0<Q_i/Q_o≤0.025$. The distance along the flow direction between the edges of adjacent droplets was given by $f=l-d_n/2$ (m=1). Figure 6(b) shows that the first droplet formation transition occurred when $0.1<Q_i/Q_o<0.5$. The droplet shape was discoid, and the droplets formed a double-layer dispersion arrangement. Thus, $f=l-d_n/2$ (m=3). Figure 6(c) shows that the second droplet formation transition occurred when $2<Q_i/Q_o<5$, and the droplets formed a four-layer dispersion arrangement. However, the droplets were obviously affected by squeezing. Therefore, $m=4$, and $f=l-d_n/4$.

Detailed information on the different droplet pattern formations for different outlet channels widths (250, 300, 350, and 400 $\mu m$) are shown in Table I.

FIG. 6 Droplet pattern formation in FFD-4 as a function of $Q_i/Q_o$ with the outlet channels width $W_d$ of 400 $\mu m$. (a) 0.025, (b) 0.15, and (c) 2. The scale bar is 200 $\mu m$. 

increased until it equaled 0 ($Q_i/Q_o=0.1$). Figure 5(b) shows that the first droplet formation transition appeared when $1<Q_i/Q_o≤3.5$. The droplet shape was discoid and resulted in a three-layer dispersion arrangement ($m=3$). Therefore, $f=l-d_n/3$. No obvious squeezing phenomenon was observed. The value of $f$ decreased until it was reduced to zero.

Two transitions of the droplet pattern formation appeared. When $Q_i/Q_o$ was large, $C_n$ was large and the shear forces on the droplet were sufficiently large to overcome the surface tension. Droplets formed a double-layer and monolayer squeezing arrangement. Figure 5(c) shows that the second droplet formation transition appeared when $3.5<Q_i/Q_o<4$, and the droplets formed a double-layer squeezing arrangement. However, the droplets appeared squeezed and in long strips. Figure 5(d) shows that the third droplet formation transition appeared when $Q_i/Q_o≥4$, and the droplets formed a monolayer squeezing arrangement. Meanwhile, the droplets became long strips.
IV. CONCLUSION

A new detailed strategy was provided to control drop size and drop pattern formation to emulsification in the microfluidic system. The microfluidic system exhibited a laminar flow with a low Reynolds number. The droplet pattern formation appeared to be a competition between shear forces and surface tension in the microchannels. Considering that geometric effects and relative fluid are significant in pattern formation, two types of emulsification processes were studied by adjusting the flow rate ratio and changing the outlet channel widths. We observed droplet patterns that ranged from monolayer dispersion to multilayer and monolayer squeezing arrangement. However, additional work is required to conduct stringent tests of theoretical models in the microfluidic system. These results provide a theoretical guidance to synthesize dispersion or polydisperse colloid particles. The system can also be applied as a component of a microfluidic screening chip.

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