Hydrodynamically mediated breakup of droplets in microchannels

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A flow focusing junction is integrated into a microchannel to break up droplets into a controllable number and size of daughter droplets. High speed images of the breakup at the flow focusing junction show that the breakup depends on the interplay between interfacial tension, shear force on the interface, and the confinement of the microchannel. Phase diagrams of the splitting performance show that the breakup is controllable by varying the flow rate of the continuous phase or by varying the size of the mother droplet. © *2011 American Institute of Physics*. [doi:10.1063/1.3552680]

Droplets in microfluidic devices can serve as vessels for chemical reaction.[1] Compared with continuous-flow microfluidics, droplet-based microfluidics have many advantages. Recirculation formed within the droplet[2] allows mixing and reaction to be achieved in milliseconds.[3] Residence time[4] and cross contamination can be significantly reduced. In the past decade, droplet-based functions such as mixing,[5] microreactors,[3] and logic operation have been developed.[6] In contrast to the bulk emulsification process that produces polydispersed droplets, microfluidic devices can generate monodispersed droplets.[7,8] Different manipulation schemes are available for the formed droplets, such as fusion,[9] splitting,[10] sorting,[11] trapping,[12] and storage.[13] Although many active means have been explored,[9,11,14] reliable passive methods without moving parts are more favorable as they are more robust and can be easily integrated without using external actuators. The experimental capacity of a droplet-based analysis can be scaled up by splitting a droplet into two or more daughter droplets. After splitting, the concentrations of the reagent in all daughter droplets are identical and suitable for subsequent parallel processes. To split droplets in a microchannel, a bifurcation junction can be used to assist the breakup of droplets.[10] At a bifurcation, the droplet is split either symmetrically or asymmetrically depending on the downstream flow resistance. Asymmetric splitting can be controlled by heat[15] or obstacles.[10]

In this letter, we propose a droplet splitting method based on hydrodynamic focusing. This method provides a way to further adjust the size of a droplet after its formation. In contrast to splitting with bifurcation or obstacles, the droplet is divided along its length in the flow direction. The number and the size of the daughter droplets can be tuned by changing the splitting flow rate of the continuous phase without changing the geometry of the microchannel. A bifurcation can only form two daughter droplets. To investigate the breakup process of droplets based on hydrodynamic focusing, water droplets in oil were formed at a flow focusing junction (Fig. 1). The microfluidic chip was fabricated in polydimethylsiloxane (PDMS, Sylgard 184, Dow Corning, Midland, MI, USA). The PDMS channels were immediately bonded with another piece of flat PDMS surface after being treated with oxygen plasma. Deionized (DI) water and mineral oil (Sigma M5904, St. Louis, USA) were used as the dispersed phase and the continuous phase, respectively. Nonionic surfactant Span 80 (Sigma S6760) 2% by weight was added into the mineral oil to assist the formation of droplets and to stabilize them against coalescence. Fluids were introduced at each inlet by syringe pumps (KD scientific, Hollistan, MA, USA) to control the flow rates. A high speed camera (Photron FASTCAM APX RS, San Diego, CA, USA) was attached to an inverted microscope (Nikon ECLIPSE TE2000-E, Tokyo, Japan) to capture the images of the droplets. The high speed images were captured at a frame rate of 250 or 500 fps and a resolution of 1024 $\times$128 pixels and were subsequently processed with a MATLAB program.

The typical process of splitting a droplet is shown in Fig. 2. A droplet in the form of a plug arriving at the flow focusing junction undergoes five stages to produce the first daughter droplet: Bulging [Fig. 2(b)], blocking [Fig. 2(c)], squeezing/stretching [Fig. 2(d)], breaking [Fig. 2(e)], and recovering [Fig. 2(f)]. After the breakup of the first daughter droplet, if the remaining droplet is sufficiently large, it can be further split by the focusing flow. Due to the lack of time for shape recovery, the remaining droplet has a slender head, facilitating the instability for the subsequent breakup. The second daughter droplet is therefore smaller than the first daughter droplet [Fig. 2(i)]. The breakup process repeats itself until the remaining droplet is not large enough for further breakup. The final droplet passes the junction without breakup [Fig. 2(j)].

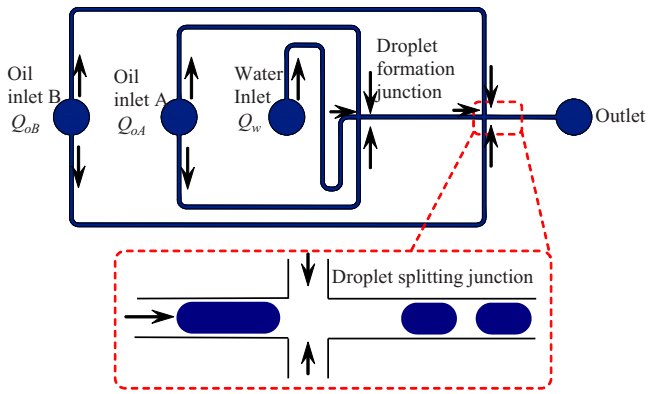


FIG. 1. (Color online) Schematics of the microfluidic device. Droplets are formed at the first junction (droplet formation junction) and then split at the second junction (droplet splitting junction). The width and height of the channel are 100 and 70 μm. The oil inlet B is used for droplet splitting.

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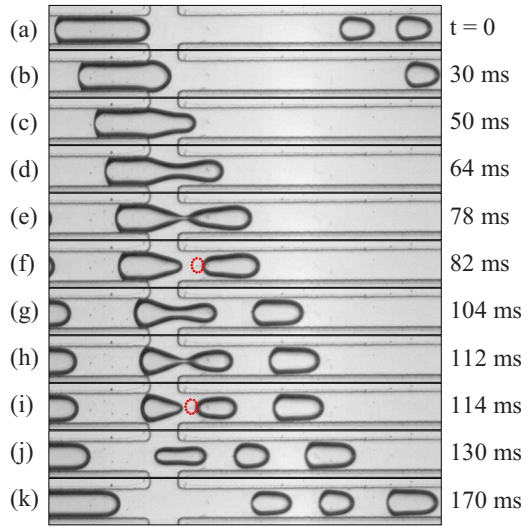


FIG. 2. (Color online) Sequences of a typical breakup process at the flow focusing junction. The flow rate are, respectively, $Q_w:Q_{oA}:Q_{oB}=40:40:140$ μL/h. Satellite droplets are highlighted with dashed circles in (f) and (i). (a) The mother droplet arrives at the junction. (b) Bulging of the head of the mother droplet. (c) The head partially blocks the downstream channel. (d) The mother droplet is squeezed and stretched. (e) The waist shrinks. (f) The detached daughter droplet recovers its plug shape. (g) The remaining droplet is squeezed and stretched. (h) The waist shrinks to form the second daughter droplet. (i) The second daughter droplet recovers its plug shape. (j) The remaining droplet passes the junction without breakup. (k) The three daughter droplets of plug shapes flow downstream.

The splitting performance was studied by varying the flow rate of the oil (Q_{oB}) and by varying the mother droplet size. The splitting flow rate of Q_{oB} was varied while the flow rates at other inlets (Q_w and Q_{oA}) were fixed. Therefore, different splitting flow rates were applied on identical droplets. For small splitting flow rates such as $Q_{oB}=50$ μL/h (Fig. 3), the droplet passes through the flow focusing junction without producing daughter droplets. As the splitting flow rates Q_{oB} increase, the squeezing and stretching force of the splitting oil also increases. When the splitting flow rate Q_{oB} is larger than the first critical value ($Q_{oB,c1}=60$ μL/h), the squeezing force is sufficiently large and is able to squeeze and stretch the waist of the droplet. The droplet subsequently breaks up into two parts. When the splitting

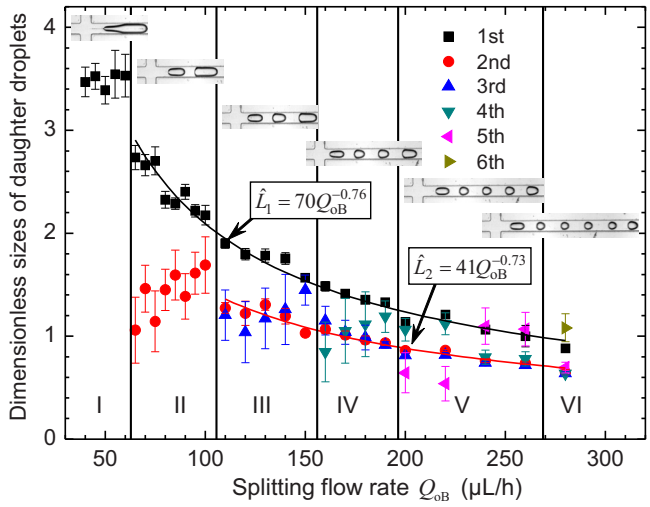


FIG. 3. (Color online) Regimes of the droplet splitting performance for different flow rates.

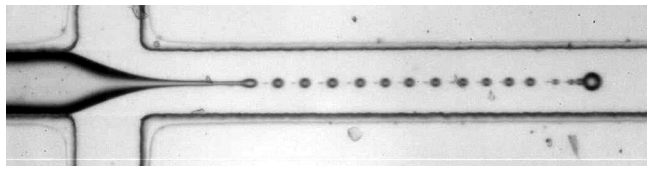


FIG. 4. Breakup of droplet in the jetting regime ($Q_w:Q_{oA}:Q_{oB}=40:40:1000$ μL/h).

flow rate Q_{oB} further increases, the squeezing and stretching effects need less time to develop the instability. The location of the surface instability shifts toward the head of the mother droplet, resulting in a smaller daughter droplet. The remaining droplet after the first breakup is large enough to form a second daughter droplet at a second critical splitting flow rate of ($Q_{oB,c2}=100$ μL/h). Increasing the splitting flow rate further, more daughter droplets can be produced (Fig. 3). The size of the last droplet is determined mainly by the amount of the remaining fluid after the previous breakup. The splitting process shares some common features with the continuous droplet formation at a flow focusing geometry. For the cases of obtaining two or more daughter droplets, the relationship between the sizes of the first daughter droplet and the splitting flow rates Q_{oB} is fitted by the least square method as

$$\hat{L} = \alpha Q_{oB}^{-\beta}. \quad (1)$$

We obtained $\hat{L}_1=70Q_{oB}^{-0.76}$ (Fig. 3). The size of the second daughter droplet was also fitted to $\hat{L}_2=41Q_{oB}^{-0.73}$. The fitting of the curve across many regimes indicates that the formation of the daughter droplet at the early stage is not significantly affected by the formation at the later stage. At a higher flow rate Q_{oB}, the breakup of the droplet will shift to a jetting regime where the breakup process happens at the end of a long column of liquid downstream of the flow focusing junction (Fig. 4). In this regime, the size of the daughter droplet is smaller than the width of the microchannel. The droplet breakup is dominated by Rayleigh capillary instability, and it appears to be less controlled.[16]

To investigate the effect of the size of the mother droplet on the splitting process, the flow rates to produce mother droplets (Q_w and Q_{oA}) were varied while maintaining a constant total flow rate (Q_w+Q_{oA}). The flow rate of droplet splitting Q_{oB} was also fixed to apply a constant focusing flow to droplets of different sizes. The regimes of droplet splitting performance for mother droplets of different sizes are shown in Fig. 5. After passing the formation junction, the mother droplet can break into daughter droplets of different numbers and different sizes. When passing the splitting junction, small droplets do not break up because they are stiff against squeezing and are hard to deform. When the mother droplet exceeds a critical size ($\hat{L}_{c1}=2.35$ in our experiment), the splitting oil has sufficient time to squeeze and stretch the droplet, leading to instability. Consequently, two daughter droplets are formed, with the second daughter droplet being smaller than the first. As the size of the mother droplet further increases, the second daughter droplet gradually increases in size while the size variation of the first daughter droplet is insignificant. If the remaining droplet is sufficiently large, the focusing flow is able to form further daughter droplets. Although this scenario is similar to the breakup of the first daughter droplet when the droplet size increases,

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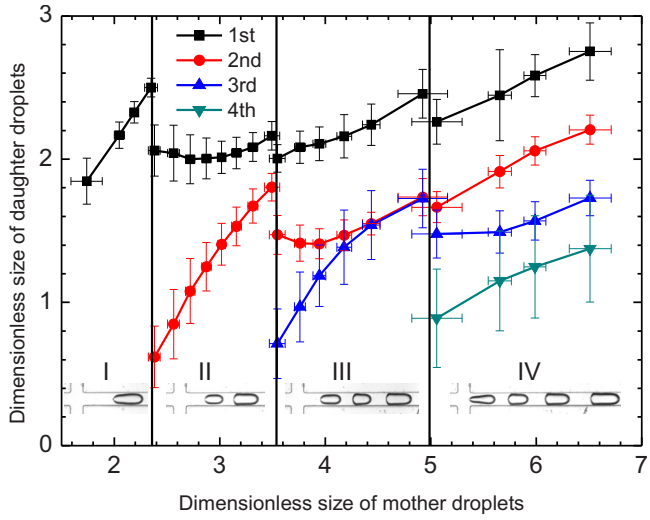


FIG. 5. (Color online) Regimes of the droplet splitting performance for different sizes of the mother droplet.

it has some unique features. As shown in Fig. 5, after the breakup of the first daughter droplet, the remaining droplet does not need to reach $\hat{L}_{c1}$ for further splitting. From Fig. 5 we can observe that the critical size for the second breakup is $\hat{L}_{c2}=1.80$. This phenomenon can also be observed for the third breakup (Fig. 5). The critical size for the third breakup is $\hat{L}_{c3}=1.75$. The critical size of the remaining drop becomes smaller because the squeezing pressure for the subsequent remaining drop is larger. The larger pressure is caused by the preceding droplet blocking the channel. The flow rates of Q_{oA} and Q_w determine the formation of the droplet upstream and also contribute to the squeezing pressure. Thus, a higher Q_{oA} and Q_w would also lead to the decrease of the critical length.

In conclusion, we investigated the breakup of droplets mediated by hydrodynamic focusing. When passing a flow focusing junction, a mother droplet can be split into two or more daughter droplets. The number and the size of the daughter droplets depend on the splitting flow rate and the size of the mother droplet. The splitting process was recorded and analyzed by high speed photography. The splitting performance was investigated by the number and the size of the daughter droplets. More droplets of smaller sizes were obtained by applying a high splitting flow rate. Alternatively, large mother droplets could also produce more daughter droplets without changing the splitting flow rates. With the flow focusing junction to split droplets in microchannels, the robustness, reliability, and flexibility to control the number and size of the daughter droplets will make it a useful tool for various droplet-based microfluidic applications.

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