Droplet Formation by Confined Liquid Threads inside Microchannels

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Supporting Information

ABSTRACT: A confined liquid thread can form monodisperse droplets near the exit of a microchannel, provided the continuous phase is able to enter the microchannel. A general model that accurately predicts the droplet size including the breakup position inside the microchannel is presented and is verified with experimental observations; breakup occurs as long as the capillary number ($Ca$) of the liquid thread is below a critical capillary number ($Ca_{cr}$); for cylindrical microchannels, it is derived that $Ca_{cr} = 1/16$. Below $Ca_{cr}$, the liquid thread remains stable and the formed droplets grow indefinitely large. The presented controlled droplet generation method is a useful tool for producing monodisperse emulsions and has great potential for the food and pharmaceutical industry.

INTRODUCTION

In recent years, microfluidic devices have been progressively used to transform both free surface and confined liquid threads in more or less monodisperse droplets with techniques based on cross-flow or co-flowing fluids such as in flow-focusing systems. In these devices, a liquid thread is actively contacted with a second immiscible fluid near a breakup region where the formation of droplets is initiated. The droplet size is directly dependent on the flow rate of the continuous phase that applies shear on the liquid thread or applies drag on the forming droplet.

Methods in microfluidics in which the flow of the continuous phase plays an insignificant role in the breakup process are terrace-based microchannel emulsification and long microchannels with a rectangular cross section. Microchannel emulsification can be scaled up by using membranes that have many thousands of separate parallel microchannels on a square centimeter of membrane area. The instability and the ultimate breakup of the confined liquid thread in microchannels are driven by a mismatch between the Laplace pressure of the growing droplet at the exit of the microchannel and of the liquid thread inside the microchannel. Sometimes, breakup is hampered by external forces, such as buoyancy, leading to nonuniform droplet sizes. An analytical model to describe the physics behind the breakup inside a microchannel, in particular the location where the breakup occurs, is still lacking. By neglecting the pressure gradient within the dispersed phase, a model to predict droplet sizes in a quasi-static regime has been described. Also, it was observed that droplets grow larger at higher flow rates and eventually grow indefinitely at very high flow rates. Here, we report a model to describe the instability and breakup of a confined viscous liquid thread inside a microchannel based on a circular cross section, which can be extended to microchannels with other geometries. Such a model that provides an insight into the parameters which determine the breakup of the droplet and predicts the size of the corresponding droplets and the location inside the microchannel where the breakup occurs has not been presented before. This paper describes for the first time the exact relationship between the droplet size and the microchannel diameter and describes the transition between making droplets with a finite diameter and making droplets with an infinite diameter. In addition, the location of the breakup inside the microchannel is predicted by considering in detail the pressure gradient of the dispersed phase inside the microchannel. Moreover, all presented relationships are solely determined by the following physical observables: interfacial tension ($\gamma$), viscosity of the dispersed phase ($\eta$), velocity of the dispersed phase ($v$), droplet radius ($R_d$), microchannel radius ($R_c$), and location of the breakup inside the microchannel ($H$). The model is experimentally verified by studying the breakup inside a microchannel in which corrugations are introduced to enable passive inflow of the continuous phase to initiate the breakup process (Figure 1). Because of the interfacial tension, the liquid thread will have a cylindrical shape with a radius $R$, comparable with the radius of the microchannel. This corrugated microchannel together with a fully controlled experimental setup allowed us to study in detail the growth of the droplets. Our findings provide new insights into the breakup process and will bring understanding of the droplet formation to a next level.

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MATERIALS AND METHODS

Silicone oil DC 200 fluid 350 Cs (Dow Corning) and silicone oil AS100 (Fluka) were used as the dispersed phase. Demineralized water containing the surfactant Tween 20 (Merck) (1 and 5 w/v %) or sodium dodecyl sulfate (Fluka) (1 w/v %) was used as the continuous phase. Viscosities were measured using a rheometer (Paar Physica MCR 300). Interfacial tensions have been measured using the pendant drop method from droplet profile analysis. Viscosities and interfacial tensions are listed in Table 1.

The velocity of the continuous phase outside the microchannel was set lower than 1 mm/s to prevent premature breakup of the liquid thread by viscous drag of the continuous phase exerted on the forming droplet. After breakup of the liquid thread, the formed droplets at the exit of the microchannel were removed to clear the view of the droplet formation process.

A microfluidic setup (Figure 1a) was placed under an optical microscope (Olympus BH2) connected to a Motic MC 2000 camera (about 15 frames per second (fps)). Data analysis was performed with computer software ImageJ (NIH) and MATLAB (MathWorks). The dispersed phase was pressed through the microchannel using a pressurized nitrogen-controlled Wallace & Tiernan device. The dispersed phase flow rate was generated by a constant pressure without any pulsations to prevent premature breakup. Typical Reynolds numbers for a laminar flow ($Re < 0.001$) are employed to yield a fully developed viscous laminar flow inside the microchannel. Typical Weber numbers are very small ($We < 10^{-5}$); therefore, inertial effects can be neglected during the breakup process. All the microchannels were designed and made in-house by inserting nine small prestretched oval-shaped glass capillaries inside a larger glass capillary with an outer diameter of 300 μm. The resulting inner star-shaped round cross section has an inner radius ($R_c$) of 50 or 62 μm (Figure 1b). Both dimensions revealed the same behavior during the experiments.

RESULTS AND DISCUSSION

Phenomenon of Liquid Thread’s Instability inside Microchannels Driven by Laplace Pressure. The process of Laplace pressure-driven breakup inside the round-corrugated microchannel is depicted in Figure 2a−d (movie is provided in Supporting Information 1). The dispersed phase is slowly pressed through the microchannel (Figure 2a). When the liquid reaches the exit of the microchannel, it will grow to a small droplet (Figure 2b). At the moment when the radius of the droplet is about two times bigger than the inner radius ($R_c$) of the microchannel, the liquid thread develops an indentation or instability (Figure 2c). Collapse of the liquid thread can be subsequently observed inside the microchannel when the instability is amplified. Two half spheres are formed after the breakup: one part attached to the droplet flows into the droplet

Figure 1. (a) Schematic representation of the microfluidic setup. (b) Cross section of a star-shaped corrugated round microchannel with nine corrugations.

Table 1. Physical Properties of the Dispersed and Continuous Phase: Interfacial Tension $\gamma$ and Viscosity $\eta$

| oil type        | dispersed phase $\eta$ (mPa s) | dispersed phase $\gamma$ (mN/m) | continuous phase $\eta$ (mPa s) |
|-----------------|-------------------------------|---------------------------------|---------------------------------|
| silicone oil    | 110                           | 1% Tween 20 [4.0, 3.5]          | 0.9                             |
| silicone oil    | 370                           | 5% Tween 20 [5.0, 4.5]          | 0.9                             |

Figure 2. A sequence of snapshots and their drawings showing the breakup of the dispersed phase thread and the formation of a droplet at the exit of a round-corrugated microchannel. (a) Liquid thread of the dispersed phase is led through the microchannel; the front of the thread forms a spherical shape. (b) Droplet is formed at the exit of the microchannel. (c) Instability is formed visible as an indentation of the liquid thread. (d) Collapse of the liquid thread inside the microchannel. Continuous phase partially replaces the dispersed phase via the corrugations of the microchannel.
and the other part stays connected to the liquid thread (Figure 2d).

**Model Description.** According to the proposed model, the breakup of the confined liquid thread will take place when two criteria are simultaneously fulfilled: an energy criterion (3.2.1) and a pressure criterion (3.2.2).

**Energy Criterion.** During the breakup process, the change of the Gibbs energy in the two fluid system \( (\Delta G = \gamma \Delta A + p \Delta V) \) will be negative. The term \( \Delta V \) is zero because both the dispersed and continuous phases are considered incompressible. Interfacial tension \( (\gamma) \) is assumed to be constant. Therefore, the total interfacial area \( (\Delta A) \) between the dispersed and the continuous phase will decrease during the breakup process.

During the breakup process of the liquid thread, a cylinder of the dispersed phase with a length \( H \) and a radius \( R_c \) with a total interfacial area \( \Delta A_t = 2\pi R_c H \), will be displaced (Figure 2b, right). The remaining surface will form two half-spherical caps after the collapse of the cylinder, and we estimate that each cap surface has an interfacial area \( \Delta A_c = 2\pi R_c^2 \) (Figure 2d, right). The reduction in the interfacial area \( \Delta A_c \) before and after the breakup is therefore

\[
\Delta A_c = 2\Delta A_2 - \Delta A_1 = 4\pi R_c^2 - 2\pi R_c H
\]

The displaced dispersed phase volume is taken up by the droplet, which will therefore increase in volume and interfacial area \( (\Delta A_d) \). The total amount of the interfacial area loss is \( \Delta A \) and should be negative

\[
\Delta A = \Delta A_c + \Delta A_d = 4\pi R_c^2 - 2\pi R_c H + \Delta A_d < 0
\]

The corresponding increase in volume of the droplet \( \Delta V_d \) is

\[
\Delta V_d = \pi R_c^2 H - \frac{4}{3} \pi R_c^3
\]

**Pressure Criterion.** Initially, the dispersed phase is pressed against the corrugated wall along the microchannel. When the droplet at the exit grows twice as big as the radius of the microchannel, the Laplace pressure of the droplet becomes equal to that of a free surface liquid thread with the same radius as that of the microchannel. Upon further growth, the dispersed phase near the exit will no longer be pressed against the wall (Figure 2c). The liquid thread will then have a free surface part near the exit and may become unstable. This means that the liquid thread then develops a neck with a local negative pressure. The first term on the right is the contribution by Hagen–Poiseuille flow through the microchannel, where \( \eta \) is the viscosity of the dispersed phase. The second term is the Laplace pressure of the droplet as determined by eq 4. When the droplet at the exit has grown to twice the diameter of the microchannel, the Laplace pressure of the droplet becomes equal to the Laplace pressure of the microchannel, the dispersed phase near the exit will no longer be pressed against the wall, and the fluid thread will become unstable. Breakup is thus initiated when at a specific breakup point \( z = H \) inside the microchannel

\[
P_{z=H} \cong \frac{\gamma}{R_c}
\]

Combining eqs 5 and 6 yields a fairly simple but general relationship between \( R_d \) and \( R_c \) at the onset of breakup

\[
\frac{R_d}{R_c} = 2 \left( \frac{C_{a_t}}{C_{a_c} - C_a} \right)
\]

with \( C_a \) as the capillary number

\[
C_a \equiv \frac{\eta v}{\gamma}
\]

and \( C_{a_t} \) is defined as the critical capillary number

\[
C_{a_{t}} \equiv \frac{1}{8} \frac{R_c}{H}
\]

Equation 7 predicts that the radius of the droplet \( R_d \) becomes indefinitely large as soon as \( C_a \) reaches the value of \( C_{a_t} \).

**Combined Result of the Energy Criterion and Pressure Criterion.** The result of the pressure criterion (eq 7) can be combined with that of the energy criterion (eqs 2 and 3). The radius of the droplet at the onset of breakup is denoted as \( R_{d0} \), and the radius of the final droplet after breakup is denoted as \( R_{di} \). The final droplet volume \( V_{di} \) can then be calculated as

\[
V_{di} = \frac{4}{3} \pi R_{di}^3 = \frac{4}{3} \pi R_{d0}^3 + \pi R_c^2 \left( H - \frac{4}{3} R_c \right)
\]

The radius of the final droplet \( R_{di} \) after the breakup can be expressed by using eq 7 for \( R_{di} \)

\[
\frac{4}{3} \pi R_{di}^3 = \frac{4}{3} \pi \left[ 2R_c \left( \frac{C_{a_t}}{C_{a_c} - C_a} \right)^3 \right] + \pi R_c^2 \left( H - \frac{4}{3} R_c \right)
\]

Bringing \( R_{di}/R_c \) to the left side of the equation becomes

\[
\frac{R_{di}}{R_c} = \left( \frac{3}{4} + 8 \left( \frac{C_{a_t}}{C_{a_c} - C_a} \right)^3 - 1 \right)^{1/3}
\]

The increment of the surface area of the droplet can be calculated from the outflow of the dispersed phase

\[
\Delta A_d = 4\pi R_c^2 \left( \frac{3}{4} + 8 \left( \frac{C_{a_t}}{C_{a_c} - C_a} \right)^3 - 1 \right)^{2/3}
\]

\[
- \left( \frac{C_{a_t}}{C_{a_c} - C_a} \right)^2 \left( \frac{3}{4} + 8 \left( \frac{C_{a_t}}{C_{a_c} - C_a} \right)^3 - 1 \right)^{1/3}
\]
Finally, an implicit relationship for \( H \) is obtained by combining this result with eq 2 and setting the difference in interfacial area \( \Delta A \) to be zero. Breakup starts as soon as both the energy and pressure criteria are fulfilled:

\[
\frac{H}{R_c} = 2 \left( 1 + \frac{3 H}{4 R_c} + 8 \left( \frac{C_a \omega}{C_{a trim} - C_d} \right)^3 - 1 \right)^{2/3}
\]

\[
- 4 \left( \frac{C_a \omega}{C_{a trim} - C_d} \right)^2
\]

(eq 14)

\( H \) is fully determined by eq 14. The other parameters in eq 14 are all known physical parameters: \( \gamma, \eta, \nu \) (via \( C_a \)), and \( R_c \). The plots of the radius of the droplet \( R_d \) normalized by \( R_c \) as a function of \( C_a \) according to eq 12 and the breakup point \( H \) normalized by \( R_c \) as a function of \( C_a \) according to eq 14 are shown in Figure 3. Note that \( R_d \) is the radius of the formed droplet when present in the continuous phase, whereas \( R_c \) is denoted as the radius of the droplet connected to the exit of the microchannel at the onset of breakup. The breakup point \( H \) is found to be slightly dependent on \( R_c \). At a very low velocity of the dispersed phase (\( C_a \approx 0 \)), the ratio of \( H/R_c \) is 2.641. At a higher velocity (\( C_a \approx C_{a trim} \)), the ratio of \( H/R_c \) is 2, and with eq 9 this yields \( C_{a trim} = 1/16 \) is 0.0625 (eq 9).

Model Verification. In our experiments, the viscosity ratio between the dispersed phase and the continuous phase was set large (\( > 100 \)). A low viscosity of the continuous phase enables sufficient fast inflow via the corrugations toward the liquid thread to fill the space that is needed to enable the breakup. It is known that a viscosity ratio up to 1 may give similar results.9,17 To prevent the adhesion of a formed droplet at the exit of the microchannel, a mild co-flow of the continuous phase with a velocity of maximum 100 \( \mu m/s \) was used. The length of the breakup region \( H \) was measured from snapshots of the movies recorded during the breakup process. The breakup region is located inside the microchannel as measured from the exit of the microchannel (Figure 2d). The dependence of \( H/R_c \) on \( C_a/C_{a trim} \) according to eq 14 is shown in Figure 4 (solid lines). It is found that the measured values seem to concord with the theoretical ones.

The ratio between the radius of the droplet \( R_d \) and the radius of the liquid thread \( R_c \) as a function of the capillary number \( C_a \) is depicted on a logarithmic scale in Figure 5. For the values of \( C_a < 0.03 \), the droplets are about two times bigger than \( R_c \). For the values of \( 0.03 < C_a < 0.0625 \) \( (C_{a trim}) \), the neck of the liquid thread which is formed during the breakup process becomes stable, thus the droplet keeps growing to a very large droplet. From this graph, we can also predict the existence of large droplets which can grow indefinitely when \( C_a \) exceeds the value of 0.0625 \( (C_{a trim}) \). No breakup of the liquid thread as well as detachment from the inner wall of the confined liquid thread is observed (Figure 5b, and a movie is provided in Supporting Information 2).

It is realized that the pressure at the exit of the microchannel may change because the radius of the neck of the liquid thread becomes smaller than \( R_c \). This may lead to an increment in the local velocity and may alter the pressure in the neck. A substantial part of the liquid thread with a length \( L \) will then become a free surface liquid thread instead of a confined liquid thread.

The breakup of a confined liquid thread can be further generalized. Equation 6 states that breakup of the confined liquid thread will be initiated whenever the dispersed phase...
pressure of the liquid thread becomes smaller than the Laplace pressure \( p_2 < \gamma / R_s \). A value of \( z \) within \( 2 < H / R_s < 2.641 \), depending on the velocity of the dispersed phase (cf. \( Ca \)), was found for stable liquid threads. When the Laplace pressure of the large droplet can be neglected, the pressure inside the liquid thread decreases about \( \gamma / R_s \) over an axial distance \( H \). The condition for breakup of the confined liquid thread can therefore be generalized by stating that a liquid thread connected to a large droplet is stable whenever the pressure gradient inside the liquid thread near the droplet has a minimum value of \( \gamma / H = \gamma / R_s H \approx \gamma / 2R_s^2 \). When the pressure gradient drops below \( \gamma / 2R_s^2 \), the breakup of the confined liquid thread may be initiated near the connected droplet. The breakup process may even apply for a low viscosity fluid such as gas. The flow-limiting factor will then probably be the passive inflow of the continuous phase liquid through the exit of the microchannel. A more detailed experimental and simulation study is then required to further characterize the dynamics of confined air bubbles inside microchannels.

The star-shaped microchannel was found to allow stable production of monodisperse droplets because it enables the inflow of the continuous phase in contrast to a pure spherical cross section. It should be noted that other shapes for the cross section of the microchannel might work as well. For example, a star shape with a less number of arms, a rectangular shape, a triangular shape, and other nonspherical shapes also allow passive inflow of the continuous phase. The star shape with multiple arms has the advantage that the inner liquid thread (with a cylindrical cross section) will not have much direct fluidic contact with the surface of the star-shaped nozzle. The model is probably also applicable to other microchannel methods of monodisperse droplet production, as long as the continuous phase is able to enter the edge or exit of the microchannel to enable the breakup process.

The interfacial tension might considerably vary during the droplet formation because of the time required for adsorption and transport of surfactants to the interface. Fast lateral transport of surfactant molecules along the interface will occur because of the Marangoni effect. This implies that the interface will exhibit a quasi-static uniform interfacial tension value, whose absolute value is determined by the amount of surfactant molecules adsorbed at the interface. It was observed that the droplet size depends on the surfactant concentration. At concentrations of Tween 20 lower than 0.5%, the droplet size became slightly larger but hardly changed at higher concentrations (1 and 5%).

**CONCLUSIONS**

A unique way to produce uniform droplets via a Laplace pressure-driven instability inside a corrugated cylindrical microchannel is demonstrated. An analytical model predicting accurately the radius of the droplet \( R_d \) as a function of the capillary number \( (Ca) \) of the dispersed phase is described. The model is well-supported by the experimental data. A value of 1/16 for the critical capillary number \( Ca_{cr} \) derived from the model was found to be an important parameter in droplet formation; above this value of \( Ca_{cr} \) the liquid thread remains stable and the attached droplet grows infinitely large. A phenomenon of a partly collapsed liquid thread was observed in the range of \( Ca \) 0.03–0.0625. The condition for breakup of the confined liquid thread can therefore be generalized by stating that a liquid thread connected to a large droplet is stable whenever the pressure gradient inside the liquid thread near the droplet has a minimum value of \( \gamma / R_s / H = \gamma / R_s H \approx \gamma / 2R_s^2 \).

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.langmuir.7b01668.

- Movie of an example of instability of a confined liquid thread (AVI)
- Movie of an example of incomplete instability of a confined liquid thread (AVI)

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**Notes**

The authors declare no competing financial interest.

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**ABBREVIATIONS**

- \( Ca \), capillary number; \( Ca_{cr} \), critical capillary number; SDS, sodium dodecyl sulfate; \( WE \), Weber number

**REFERENCES**

1. Eggers, J.; Villenauex, E. Physics of Liquid Jets. *Rep. Prog. Phys.* 2008, 71, 036601.
2. Seemann, R.; Brinkmann, M.; Pföh, T.; Herminghaus, S. Droplet Based Microfluidics. *Rep. Prog. Phys.* 2012, 75, 016601.
3. Basaran, O. A. Small-Scale Free Surface Flows with Breakup: Drop Formation and Emerging Applications. *AIChE J.* 2002, 48, 1842–1848.
4. Steegmans, M. L. J.; Schroën, K. G. P. H.; Boom, R. M. Characterization of Emulsification at Flat Microchannel Y Junctions. *Langmuir* 2009, 25, 3396–3401.
5. Peng, S. J.; Williams, R. A. Controlled Production of Emulsions Using a Crossflow Membrane: Part I: Droplet Formation from a Single Pore. *Chem. Eng. Res. Des.* 1998, 76, 894–901.
6. Ushikubo, F. Y.; Birrelli, F. S.; Oliveira, D. R. B.; Cunha, R. L. Y- and T-Junction Microfluidic Devices: Effect of Fluids and Interface Properties and Operating Conditions. *Microfluid. Nanofluid.* 2014, 17, 711–720.
7. Christov, N. C.; Danov, K. D.; Danova, D. K.; Kralchevsky, P. A. The Drop Size in Membrane Emulsification Determined from the Balance of Capillary and Hydrodynamic Forces. *Langmuir* 2008, 24, 1397–1410.
8. Umbanhowar, P. B.; Prasad, V.; Weitz, D. A. Monodisperse Emulsion Generation via Drop Break off in a Coflowing Stream. *Langmuir* 2000, 16, 347–351.
9. Utada, A. S.; Fernández-Nieves, A.; Stone, H. A.; Weitz, D. A. Dripping to Jetting Transitions in Coflowing Liquid Streams. *Phys. Rev. Lett.* 2007, 99, 094502.
10. Castro-Hernández, E.; Gundabala, V.; Fernández-Nieves, A.; Gordillo, J. M. Scaling the Drop Size in Coflow Experiments. *New J. Phys.* 2009, 11, 075021.
(11) Garstecki, P.; Stone, H. A.; Whitesides, G. M. Mechanism for Flow-Rate Controlled Breakup in Confined Geometries: A Route to Monodisperse Emulsions. Phys. Rev. Lett. 2005, 94, 164501.

(12) Nie, Z.; Seo, M.; Xu, S.; Lewis, P. C.; Mok, M.; Kumacheva, E.; Whitesides, G. M.; Garstecki, P.; Stone, H. A. Emulsification in a Microfluidic Flow-Focusing Device: Effect of the Viscosities of the Liquids. Microfluid. Nanofluid. 2008, 5, 585–594.

(13) Sugiyama, S.; Nakajima, M.; Iwamoto, S.; Seki, M. Interface Tension Driven Monodisperse Droplet Formation from Microfabricated Channel Array. Langmuir 2001, 17, 5562–5566.

(14) Sugiyama, S.; Nakajima, M.; Seki, M. Effect of Channel Structure on Microchannel Emulsification. Langmuir 2002, 18, 5708–5712.

(15) Kobayashi, I.; Uemura, K.; Nakajima, M. Controlled Generation of Monodisperse Discoid Droplets Using Microchannel Arrays. Langmuir 2006, 22, 10893–10897.

(16) Kobayashi, I.; Neves, M. A.; Yokota, T.; Uemura, K.; Nakajima, M. Generation of Geometrically Confined Droplets Using Microchannel Arrays: Effects of Channel and Step Structure. Ind. Eng. Chem. Res. 2009, 48, 8848–8855.

(17) van Dijke, K. C.; Schröën, K. C. P. G. H.; Boom, R. M. Microchannel Emulsification: From Computational Fluid Dynamics to Predictive Analytical Model. Langmuir 2008, 24, 10107–10115.

(18) van der Zwan, E.; Schröën, K.; Boom, R. A Geometric Model for the Dynamics of Microchannel Emulsification. Langmuir 2009, 25, 7320–7327.

(19) Kobayashi, I.; Nakajima, M.; Chun, K.; Kikuchi, Y.; Fujita, H. Silicon Array of Elongated through-Holes for Monodisperse Emulsion Droplet. AIChE J. 2002, 48, 1639–1644.

(20) van Dijke, K.; Veldhuis, G.; Schröën, K.; Boom, R. Parallelized Edge-Based Droplet Generation (EDGE) Devices. Lab Chip 2009, 9, 2824–2830.

(21) Dangla, R.; Kayi, S. C.; Baroud, C. N. Droplet Microfluidics Driven by Gradients of Confinement. Proc. Natl. Acad. Sci. U.S.A. 2013, 110, 853–858.

(22) Kobayashi, I.; Mukataka, S.; Nakajima, M. Effects of Type and Physical Properties of Oil Phase on Oil-in-Water Emulsion Droplet Formation in Straight-through Microchannel Emulsification, Experimental and CFD Studies. Langmuir 2005, 21, 5722–5730.

(23) Kobayashi, I.; Mukataka, S.; Nakajima, M. Effect of Slot Aspect Ratio on Droplet Formation from Silicon Straight-through Microchannels. J. Colloid Interface Sci. 2004, 279, 277–280.

(24) van Rijn, C. J. M. Membrane Emulsification. Nano and Micro Engineered Membrane Technology; Membrane Science and Technology Series; Elsevier Publishers: Amsterdam, 2004; Vol. 10, pp 347–371.

(25) Vladisavljević, G. T.; Cobayashi, I.; Nakajima, M. Production of Uniform Droplets Using Membrane, Microchannel and Microfluidic Emulsification Devices. Microfluid. Nanofluid. 2012, 13, 151–178.

(26) Barkley, S.; Scarfe, S. J.; Weeks, E. R.; Dalnoki-Veress, K. Predicting the Size of Droplets Produced through Laplace Pressure Induced Snap-Off. Soft Matter 2016, 12, 7398–7404.

(27) Khodaparast, S.; Magnini, M.; Borhani, N.; Thorne, J. R. Dynamics of isolated confined air bubbles in liquid flows through circular microchannels: an experimental and numerical study. Microfluid. Nanofluid. 2015, 19, 209–234.

(28) Schröën, K.; Ferrando, M.; de Lamo-Castellvi, S.; Sahin, S.; Güell, C. Linking Findings in Microfluidics to Membrane Emulsification Process Design: The Importance of Wettability and Component Interactions with Interfaces. Membranes 2016, 6, 26.