Self-triggering regime for synchronized formation of two droplets

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(Dated: 13 February 2014)

This letter reports experimental results of the synchronized formation of two liquid droplets in a microfluidic device. A pair of droplets is formed periodically in a T-junction configuration with a single channel for the continuous phase and two inlets for the dispersed phase. The pair-wise droplet formation process is self-triggering, as the first droplet formed upstream triggers the breakup of the second droplet downstream. The triggered breakup happens across the different formation regimes. The effects of capillary number and flow rate ratio on the size and order of the droplets are investigated. The configuration reported here may serve as a parallel to serial sampling device for droplet-based lab-on-a-chip platforms.

PACS numbers: 47.55.df, 47.61.Fg, 47.60.-i, 47.85.Np, 47.55.pb, 85.85.+j

Keywords: Droplets, parallel to serial, T-junction, droplet-based microfluidics

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Samples for biochemical and chemical assays are commonly stored in a well plate array and transferred to subsequent analysis steps by manual or automated robotic pipetting. Advances in microfluidics and lab-on-a-chip (LOC) technologies led to improvement in sample handling for conventional analytical techniques, in particular mass spectrometry\(^1\). Using a compartmented droplet as a sample container, the volume can be reduced by several orders of magnitude to picoliters range. Droplet-based microfluidics allows the compartmentalization of aqueous samples in an immiscible oil phase\(^2\). The formation of multiple sample droplets is the first challenge of adapting droplet-based microfluidics technology to applications using conventional detection tools such as mass spectrometry. With of-the-shelf polytetrafluoroethylene (PTFE) tubings as storage and transport platform, automated robotic platforms has been used to convert samples from a micro well array\(^3,4\) or from a rotating carousel\(^5\) into a droplet train. To date, no integrated solution for the conversion of liquid samples in well array plate into an addressable droplet train.

Barbier et al. reported a system with two T-junction placed in parallel for the formation of two separate disperse phases\(^6\). The hydraulic coupling between the T-junctions show a very complex dynamic behavior. Hashimoto et al. later applied the same parallelization approach to maximum four flow-focusing junctions\(^7\) for the formation of bubbles. However, the design does not allow for the orderly lineup of the droplets. Frenz et al.\(^8\) concept for the formation of a droplet pair similar to that of Barbier et al.\(^6\). The droplets are formed in squeezing regime warranting the order of the formed droplet pair. Chokkalingam et al. reports a similar design for the formation of a droplet pair\(^9\). The synchronization was achieved by the coupling of the pressure at the two formation junctions. Hong et al. also reports another similar configuration for synchronized formation of a droplet pair with different contents\(^10\). In these previous works, the formation can only occur in the squeezing regime because the droplet should alternatively blocks one of the two branches.

This letter investigates the parallel formation of a droplet pair with different contents. The basic T-junction configuration was selected. The formation regime changes from squeezing, to dripping to parallel flow depending on the capillary number \(Ca = \frac{\mu_o U_o}{\sigma}\) and the flow rate ratio \(\alpha = \frac{Q_o}{Q_w}\), where \(\mu_o\) and \(U_o\) are the viscosity and mean velocity of oil as the carrier phase, \(\sigma\) is the interfacial tension. Since we aim to have a quick regime change and possibly to reduce the operation parameters to the flow rate ratio only, operation at capillary numbers on the order of unity is targeted\(^11\). The configuration consists of
a main channel for the continuous phase (oil) and two parallel T-junctions for the disperse phases. Because only one droplet can breakup at a time, the configuration is suitable for the self-synchronized formation of droplets with different contents. Self-synchronization is triggered by a the droplet formed upstream. The hydrodynamic interaction caused by the flow field around the upstream droplet forces the breakup of the second droplet. In turn, the breakup of the downstream droplet causes a sudden pressure drop across the forming upstream droplet and initiates the new cycle of pair-wise droplet formation. To keep the upstream droplet close to the forming downstream droplet, all inlet channels are placed on one side of the main channel. For synchronization to occur, the time for droplet breakup should be the same as the time for a droplet to travel from one inlet to the other inlet:

\[ \frac{V_d}{Q_w} = \frac{L}{U_{av}} \]  

where \( V_d \) is the volume of the droplet, \( Q_w \) is the volumetric flow rate of the aqueous samples, \( L \) is the distance between the inlets and \( U_{av} = (Q_o + Q_w)A \) is the average velocity in the main channel between the inlets with \( A \) the channel cross-sectional area. With an estimated droplet volume of \( V_d = \frac{\pi D_d^3}{6} \), the relationship between the droplet diameter and the geometric parameters as well as the flow rate ratio is:

\[ D_d = \frac{6^{\frac{1}{2}}}{\pi} A^\frac{1}{2} L^\frac{1}{2} \left( \frac{1}{\alpha + 1} \right)^\frac{1}{4} \]  

Figure 1 shows the channel design with two inlets for the disperse phases. The channels are 100 \( \mu \)m high. The main channel is 100 \( \mu \)m wide, while the side channels are 50 \( \mu \)m wide. With a square cross section, the hydraulic diameter of the main channel is the same as the channel width and height, \( D_h = 100 \mu \)m. The device consists of a polydimethylsiloxane (PDMS) part bonded onto the glass slide. Photoresist SU-8 with a thickness 100 \( \mu \)m was coated and patterned on a silicon wafer by photolithography. The PDMS device was then casted and peeled off from the SU-8 mold and bonded to the glass slide with the help of oxygen plasma treatment.

In our experiments, de-ionized (DI) water and mineral oil (Sigma-Aldrich 330760, St. Louis, USA) were used as the dispersed phase and the continuous phase, respectively. Surfactant Span 80 (Sigma S6760) was added into the mineral oil (0.5% by weight) to reduce the interfacial tension. The density of the oil is \( \rho_o = 862 \text{ kg/m}^3 \). The dynamic viscosity of the oil is \( \mu_o = 170 \times 10^{-3} \text{ Pa.s} \) (at 20 \( ^\circ \)C). The interfacial tension between oil and DI
FIG. 1. Schematic of the microfluidic device for the generation of multiple droplets with different contents: (a) Key geometric parameters (in μm); (b) Device assembly.

Water of $\sigma = 4.86 \pm 0.50$ mN/m was measured using a tensiometer at 25°C (FTA200, First Ten Angstrom). Two precision syringe pumps (KD scientific, USA). The water inlets are driven by the same pump with the same syringe size. An epi-fluorescent inverted microscope (Nikon Eclipse TE 2000-E) and a high speed camera (Photron FASTCAM APX RS, USA) was used to capture the droplet merging process at a frame rate of 500 frames per second. The recorded images were then processed by a customized MATLAB program.

We kept the flow rate of water at $Q_w = 200 \, \mu L/h$ while increasing oil flow rate $Q_o$ from 500 $\mu L/h$ to 3500 $\mu L/h$. The corresponding capillary number and Reynolds number ($Re = \rho o U_o D_h/\mu o$) range from Ca=0.486 to Ca=3.40 and Re=$7.04 \times 10^{-3}$ to Re=$4.93 \times 10^{-2}$, respectively. Since a higher oil flow rate will cause a large pressure drop and leakage, a higher
flow rate ratio between oil and water was achieved by keeping the oil flow rate at 3500 \( \mu \)L/h and reducing the water flow rate from 200 \( \mu \)L/h to 50 \( \mu \)L/h. With a constant oil flow rate, the capillary number was kept constant at 3.4. With these relatively large capillary numbers, a quick regime change can be expected\(^{11}\), leaving the operation solely controlled by the flow rate ratio \( \alpha \). Figure 2 shows the representative images of the stable formation process of the droplet pairs. Increasing flow rate ratio further allows the hydrodynamic decoupling between the inlets. However, the self-triggering mechanism still works. In general, the formation of droplet pairs using the configuration of a pair of T-junctions has three regimes: (i) squeezing regime; (ii) dripping regime with synchronized formation and correct droplet order and (iii) dripping regime with unstable droplet pair.

Figure 3 shows the equivalent droplet diameter and the period of the droplet pairs versus the flow rate ratio across the different formation regimes. The surface area of the formed droplet was evaluated using an customized MATLAB program. With an image size of 1024 pixels \( \times \) 128 pixels, the size of a pixel is 2.6 \( \times \) 2.6 \( \mu \)m. The program converts the recorded grey-scale images into a binary image and extracts the area of the droplet. The equivalent diameter was determined as the diameter of a circle with the same surface area of the recorded droplet. For each data point, 10 droplets from 5 images were evaluated. The mean value and the standard deviation of the equivalent droplet diameter were evaluated from this data set. The relationship between the equivalent droplet diameter and the flow rate ratio follows the relationship predicted in (2). At small flow rate ratios, the disperse phase form a slug that occupies the whole channel cross section. Thus, the effective diameter does not represent the actual size of the droplet. For droplet diameters less than the channel width and height (less than 100 \( \mu \)m), the droplet can be approximated by a sphere with the effective diameter. This fact explains why the relationship of (2) (depicted as the line in Fig. 3(a)) fits the experimental data better for effective diameters less than 100 \( \mu \)m. Figure 3(b) shows the measured period between the droplet pairs. The line represents the distance of 400 \( \mu \)m between the inlets. The results show that the period is consistently the same as the distance across the different regimes.

In conclusion, a droplet pair with different contents can be formed in a T-junction configuration with two inlets. The hydrodynamic interaction between the inlets allows the synchronization through self-triggering of the droplet formation process. We showed with our device configuration that self-triggering is possible across different formation regimes.
FIG. 2. Self-triggering synchronized formation of droplet pairs.
at a relatively high capillary number on the order of unity. The large capillary number makes hydrodynamic interaction the important factor for droplet formation. The formation process is therefore determined by the flow rate ratio and can be self-synchronized. The configuration investigated in this letter can be used to produced a packet of samples for subsequent manipulation and measurements in droplet-based lab-on-a-chip platforms.

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