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Thermally mediated breakup of drops in microchannels

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The authors used thermally induced surface tension gradients to manipulate aqueous droplets in microchannels. Control of the droplet breakup process was demonstrated. Droplet sorting can be achieved with temperatures above a critical value. Numerical simulation using a two-dimensional model agrees qualitatively well with the experimental results. The used control temperature of less than 55 °C shows that this active control concept is suitable for biochemical applications. Thermal control promises to be a simple and effective manipulation method for droplet-based lab on a chip. © 2006 American Institute of Physics. [DOI: 10.1063/1.2400200]

Microfluidic technology allows the development of microreactors and analysis systems with a length scale on the order of hundreds of microns. The miniaturization promises the development of high-throughput assays with small amount of reagents. This technology allows the implementation of small massively multiplexed, arrayed assays. Although droplets and flows driven by surface and interfacial tension gradients were studied over the last 100 years,^{1,2} droplet-based phenomena recently found renewed interests from the microfluidics community with a number of applications.³ Instead of handling continuous liquid flows in microchannels, chemical and biological agents are contained in microdroplets. The droplets are encapsulated by an immiscible fluid, which is also called the carrier fluid. Existing continuous-flow microfluidic technologies have been widely used for the formation and manipulation of microdroplets. These concepts depend much on liquid properties such as density, viscosity, interfacial tension, and the flow rates of the aqueous flow and the immiscible carrier flow.⁴ The inverse effect of droplet formation is droplet breakup. At a T-junction bifurcation, a droplet can be split passively into two smaller daughter droplets. The size of the daughter droplets depends on the fluidic resistance of the bifurcation branches.⁵

In the processes described above, capillary number $Ca = \mu_1 U_1 / \sigma$, viscosity ratio μ_1 / μ_2 , and the flow rate ratio $Q_1 = Q_2$ are the main control parameters, where μ , U , σ , and Q are the viscosity, velocity, interfacial tension, and flow rate, respectively.¹⁵ Subscripts 1 and 2 denote the aqueous and the carrier phases. Alternatively, formation and breakup can be affected by forces induced by external fields. The most common methods used in the past were based on electrostatic phenomena such as electrowetting^{6,7} and dielectrophoresis.⁸ Beside electrowetting, the surface tension gradient can also be induced by a temperature gradient. Brozoska *et al.*⁹ investigated the movement of a droplet on a flat surface in a temperature field. Darhuber *et al.*¹⁰ and Tseng *et al.*¹¹ recently studied the dynamic behaviors of microdrop-

lets in a temperature field of a flat plate. All the above concepts are based on a planar platform and not suitable for a continuous-flow system. Recently, Suryo and Basaran reported the control of droplet formation based on thermally induced surface tension gradients.¹² Link *et al.* reported a method of electric control of droplets in a continuous-flow platform.¹³ In this method, electrostatic forces were added to the formation and breakup processes of microdroplets. The droplets are charged with a high voltage. Subsequently, the charged droplet can be manipulated actively by electric fields induced by electrodes in the microchannels. The platform is able to carry out basic manipulation functions such as formation, merging, breakup, and sorting.

In this letter, we report another effective method for control of droplets in microchannels. In our method, the force balance is mainly controlled by the interfacial stress induced by a temperature field, which is generated by a heater. Two-dimensional numerical models support the experimental proof-of-concept results. This concept can be easily implemented and does not need high voltages, as in the case of electric control reported by Link *et al.*¹³ Operating temperatures are kept below 55 °C, thus safe for performing assays with biomolecules and cells. Following, we report sequentially the fabrication of the device prototype, the numerical model, and the experimental results for controlled droplet breakup and droplet sorting.

Figure 1 depicts the schematic concept of the microfluidic device used in our experiments. The device was fabricated in polymethyl methacrylate (PMMA). The channel structures were engraved by a CO₂ laser into the substrate and have a depth of 100 μm. The microchannel network consists of an entrance T junction for droplet formation and a T-junction bifurcation for droplet breakup or sorting. The widths of these microchannels are given in Fig. 1. The two branches at the T-junction bifurcation are symmetrical and have the same length of 8 mm. The device is sealed on top with another PMMA plate containing the fluidic access holes. Around the microchannels air gaps are machined out of the substrate with the same laser. At the air gap of the lower exit branch, a heater is wrapped around the microchannel. The heater wire is made of nickel/chromium alloy

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FIG. 1. Schematic of the microfluidic device for active control of droplets (not to scale).

(Ni80Cr20) (Good Fellow, UK). The wire has a diameter of $125\ \mu\text{m}$ and is insulated with an $8\text{-}\mu\text{m}$ -thin polyimide layer.

Cooking oil worked as the carrier fluid in our experiments. The density and viscosity were measured as $0.912\ \text{g/cm}^3$ and $5.20 \times 10^{-2}\ \text{Pa s}$ (controlled flowrate rheometer, Contraves low shear 40), respectively. De-ionized water mixed with $0.047\ \text{wt}\%$ fluorescent dye (fluorescein disodium salt $\text{C}_{20}\text{H}_{10}\text{Na}_2\text{O}_5$) was used as the aqueous fluid. Together with an epifluorescent inverted microscope and a fast digital camera, the dye helps to visualize the droplets in the micro channels. An interfacial tension of $48.2 \pm 2 \times 10^{-3}\ \text{N/m}$ between water and the oil was measured with a tensiometer (FTA200, First Ten Angstrom). A precision syringe pump (Lomir Biomedical Inc.) drives both the oil and the aqueous fluid into the device. In the following experiments, the flow rates of the aqueous fluid and the oil were kept at constant values of $Q_1 = 60\ \mu\text{l/h}$ and $Q_2 = 300\ \mu\text{l/h}$, respectively. The temperature of the channel wall was measured with a miniature thermocouple (K type) and is representative for the heater in use.

The experimental results are compared qualitatively with simulation results from a two-dimensional model, where the droplet is assumed to have a diameter of $360\ \mu\text{m}$. While recently reported works focused on the formation and breakup of droplets in a coflow configuration,^{14,15} we focus on the simulation of thermally mediated breakup of droplets in the T-junction bifurcation. The motion of both water and oil is governed by the incompressible Navier-Stokes equations. The interface between the two phases is captured using the level-set method coupled with a global mass correction scheme.¹⁶ The variable interfacial tension (effect of temperature gradient) between the two phases is modeled within the framework of continuum surface force model.¹⁷ In the case of a lower heated bifurcation branch, the increase in the fluid temperature when approaches the lower outlet induces a lower interfacial tension. Rather than calculating the interfacial tension from the temperature field by solving the energy equation, it is sufficient for the purpose of a qualitative comparison to assume that the interfacial tension decreases linearly when approaches the lower outlet. At the inlet, the velocity profile is assumed to be fully developed. The two outlets are maintained at the same pressure. No slip and no penetration conditions are applied at the wall. The governing equations are solved using the finite volume method.¹⁸

Figure 2(a) shows the process of breakup without temperature field at the T-junction bifurcation and at a room

FIG. 2. Active control of the breakup process: (a) passive breakup, (b) thermally mediated breakup, and (c) droplet sorting.

temperature of $27\ ^\circ\text{C}$. At fixed flow rates, uniform droplets were formed at the entrance T junction. When the droplet is stretched at the T junction, the thread connecting the two bulbs of the droplet becomes thinner. Because the two branches of the T-junction bifurcation are symmetrical, each droplet is split equally into two smaller daughter droplets. In the simulation, breakup of the droplet is said to occur when the thread is smaller than the size of one control volume, i.e., the resolution of the computation. Analysis of the dynamics just prior to pinch off would require accounting for the non-planar curvature of a three-dimensional drop, as in Refs. 12, 14, and 15. We define the ratio between the sizes of the droplets in the upper branch and those in the lower branch as the size ratio α . In the case of breakup without temperature field, the size ratio is $\alpha = 1$. Previously, Link *et al.* used the lengths of the branches to control the size ratio of the two daughter droplets.⁵

In our experiment, the size ratio α was controlled by activating the heater around the lower branch, as depicted in Fig. 1. Figure 2(b) shows the breakup effect induced by the temperature field. The higher temperature at the lower branch leads to a lower viscosity and a lower interfacial tension. As a result, the droplets are split at the T-junction bi-

FIG. 3. Size ratio of daughter droplets as function of the heater temperature.

furcation into a larger daughter droplet in the heated lower branch and a smaller daughter droplet in the unheated upper branch ($\alpha < 1$). Varying the temperature can precisely control the size ratio of the daughter droplets. Therefore, a size ratio between 1 and 0 ($0 \leq \alpha \leq 1$) can be achieved by varying the temperature of the heater. A similar trend is obtained from the numerical model.

If the temperature of the lower heater is increased up to a critical value, a size ratio of $\alpha=0$ can be achieved. That means the entire droplet enters the lower branch of the bifurcation. Figure 2(c) shows the sorting effect at the T-junction bifurcation with a heater temperature of 40 °C. Such a phenomenon is reproduced in the numerical simulation, as shown in Fig. 2(c). Figure 3 shows the measured relationship between the size ratio α and the temperature of the heater. The size ratio decreases from 1 (passive breakup) to 0 (sorting), while the temperature at the lower branch increases from 27 to 40 °C.

In conclusion, we have presented a novel method for controlling the breakup process of microdroplets at a T-junction bifurcation. Aqueous droplets are formed in an

immiscible carrier flow at an entrance T junction. A heater at the T junction bifurcation can control the size ratio of the daughter droplets. At a critical temperature, the entire droplet can enter into the heated branch. The improved functionality of thermal control will allow droplet-based microfluidics to have a wide range of applications. Controlled droplet breakup allows simple and precise dispensing of reagents. The sorting capability will make this concept an effective microsorter. Cells or other bioparticles can be encapsulated in a droplet and sorted using this thermal control concept. The platform reported in this letter may potentially have an impact in droplet-based microfluidics.

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