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Thermally mediated breakup of drops in microchannels

Droplet formation in microchannels under static conditions
Thermally mediated droplet formation in microchannels

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Precise dispensing of microdroplets is an important process for droplet-based microfluidics. The droplet formation by shear force between two immiscible fluids depends on their flow rates, the viscosities, and the interfacial tension. In this letter, the authors report the use of integrated microheater and temperature sensor for controlling the droplet formation process. The technique exploits the dependency on temperature of viscosities and interfacial tension. Using a relatively low heating temperature ranging from 25 to 70 °C, the droplet diameter can be adjusted to over two times of its original value. The relatively low temperature range makes sure that this concept is applicable for droplets containing biological samples. © 2007 American Institute of Physics. [DOI: 10.1063/1.2773948]

Currently, microfluidics becomes the key technology for miniaturization of chemical and biochemical analyses, high-throughput screening, and combinatorial chemistry. In conventional microfluidic systems, liquids are handled in continuous flows. In contrast to continuous-flow microfluidics, droplet-based or digital microfluidics forms, transports, and manipulates droplets in an immiscible carrier fluid. The droplets can be handled easily and accurately with a number of techniques such as electrowetting, dielectrophoresis, thermocapillarity, and magnetic actuation.2 Similar to an analog-to-digital converter in electronics, droplet formation works as the interface between continuous-flow microfluidics and droplet-based microfluidics. Furthermore, controlling drop size has been the holy grail of microfluidic applications such as ink jet printing.3 In lab-on-a-chip applications, the droplet formation works as a dispensing process for precise fluid volumes with high uniformity.

In most recent works on droplet-based microfluidics, the only way to adjust the droplet size of a given liquid system was tuning the flow rates or the flow rate ratio. Droplet formation is usually implemented by a T-junction or by flow focusing through a small orifice. Chwalek et al.4 utilized the temperature dependency of surface tension and viscosity to control the deflection of liquid microjets. Recently, Suryo and Basaran reported numerical results of droplet formation based on thermally induced effects.5 Link et al. reported a method of electric control of droplets in a continuous-flow platform.6 A relatively high voltage and the corresponding bulky voltage supply are needed for this concept, which may not be favorable for applications that require compactness and portability. Recently, our group has shown the use of temperature to control the splitting and switching behavior of microdroplets at a T-junction bifurcation.7

In this letter, we report a method for controlling the droplet formation process using a temperature sensor and a heater. Keeping all geometrical parameters as well as the flow rates fixed, the droplet size can be adjusted by controlling the temperature at the breakup location. We used a flow-focusing configuration for the droplet formation experiments.8 The flow-focusing configuration forms droplets in different regimes such as geometry-controlled formation, thread formation, dripping, and jetting as experimentally observed by Anna and Mayer9 and numerically modeled by Suryo and Basaran.10 The formation process is mainly determined by the flow rate ratio and the capillary number $Ca = \eta a / \sigma$, where $\eta$ is the dynamic viscosity of the carrier fluid, $\sigma$ is the interfacial tension, $a$ is the shear rate, and $\alpha$ is the half width of the middle stream. By approximately balancing the Laplace pressure with the shear force, the diameter of the formed droplet $D \sim \sigma / (\eta a)$ was predicted by Taylor almost 70 years ago.11 To investigate the dependency on temperature of all parameters in this relation, the droplet diameter, the viscosity, and the surface tension are normalized by their corresponding values at a reference temperature such as the room temperature of $T_0 = 25 \, ^\circ\text{C}$: $D^* (T) = D / D_0$, $\sigma^*(T) = \sigma / \sigma_0$, and $\eta^*(T) = \eta / \eta_0$. Thus, the dimensionless temperature dependence of the droplet diameter can be described as

$$D^* (\Delta T) \approx \frac{1}{Ca_0} \frac{\sigma^*(\Delta T)}{\eta^*(\Delta T)} \Delta T$$

where $\Delta T = T - T_0$ is the temperature difference and $Ca_0 = D_0 \gamma \eta_0 / \sigma_0$ is the capillary number at room temperature using the droplet diameter as the characteristic length scale. In Eq. (1), both shear rate $\gamma$ and initial droplet diameter $D_0$ are functions of flow rates and flow rate ratio.12 Since the oil in our experiment contains surfactant, the flow rate may also affect the surfactant concentration and consequently the breakup process.13 Therefore, the problem of temperature-dependent droplet formation in a flow-focusing configuration is more complex than relationship (1). To simplify the analysis, we only investigate the correlation between $D^*$ and $\eta^*$ using the experiments reported in this letter.

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For examining the dependency on temperature of the droplet formation process, a microdevice was fabricated using micromachining of glass and polydimethylsiloxane (PDMS). To start with, the temperature sensor and the microheater were patterned using photolithography and lift-off technique. The microheater and the temperature sensor were made of thin-film platinum. A titanium layer was used as the adhesion layer between glass and platinum. The microchannel network was fabricated in PDMS using soft lithography. The master mold was fabricated by photolithography of the thick-film resist SU-8 using a low-cost transparency mask. The glass wafer with the patterned microheater and temperature sensor was subsequently coated with a thin PDMS layer before being bonded to the PDMS part containing the microfluidic network. This step makes sure that all channel walls have the same properties. Bonding is achieved after treating both PDMS surfaces with oxygen plasma. All microchannels in the device have the same depth of 70 μm. The orifice size at the junction is 45 μm. All other geometric parameters of the device are given in Fig. 1. The temperature sensor was calibrated so that its resistance can be used for in situ temperature measurement. The test chip measures 1 × 1 cm².

In the experiments, de-ionized water with 0.05% w/w fluoresence dye (Sigma F6377) was used as the aqueous phase, while mineral oil (Sigma M5904) with 2% w/w surfactant span 80 (Sigma S6760). Figure 2 shows the temperature dependence of the normalized viscosity of the oil and its normalized interfacial tension to water. The lines represent the exponential fitting curves. The dependency on temperature of the normalized interfacial tension and viscosity are $\sigma^* = \exp(-0.0144\Delta T)$ and $\eta^* = \exp(-0.0344\Delta T)$, respectively. According to Eq. (1), the expected dependency on temperature of the droplet diameter is $D^* \propto \sigma^*/\eta^* = \exp(0.02\Delta T)$. This implies that the droplet diameter increases with increasing temperature.

Droplet formation with flow rate ratios of 300:50:300 and 400:100:400 μl/h was characterized at different temperatures. Using the definition of the shear rate from Ref. 9, the capillary number at room temperature of both cases are approximately $Ca_0 = 0.08$. Using $\alpha = 100 \mu$m as the characteristic length scale, the capillary numbers of these two cases are 0.20 and 0.27, respectively. According to Anna and Mayer, the droplet formation is in the geometry-controlled regime. We used an epifluorescent inverted microscope (model ECLISPE TE2000-S) with a filter set (Nikon B-2A, excitation filter for 450–490 nm, dichroic mirror for 505 nm, and emission filter for 520 nm) for observing the droplets. A sensitive interline transfer charge coupled device (CCD) camera (HiSense MKII) was used for recording the droplet images. The CCD camera has a resolution of 1344 × 1024 pixels. The temperature sensor was calibrated before the experiments. The temperature was adjusted by tuning the voltage of the heater and monitoring the resistance of the temperature sensor. Figures 3(a) and 3(b) show the typical recorded images of the formed droplets when the heater is turned off and on. The recorded images were processed by a customized MATLAB program to measure the droplet diameter.

The droplet diameters of the two cases were normalized by their nominal values at 25 °C. Figure 4 shows measured normalized droplet diameters as a function of temperature difference for two different oil-water-oil flow rate ratios. The results show that the relative change of the droplet size is a function of temperature as predicted previously. The predicted exponential relation seems to agree only at higher temperatures. The discrepancy at lower temperatures ($\Delta T < 15$ K) may have several reasons. First, the exponential fitting functions of the viscosity data and the interfacial tension data may be too coarse. Second, the data in Fig. 4 may hint to two different formation regimes with a transition at 10 K $< \Delta T < 20$ K. Third, since the two sets of data in Fig. 4 do not overlap, the droplet diameter may also depend on the flow rate ratios and surfactant concentration as mentioned above.
In conclusion, we reported the fabrication and test of a microfluidic device for thermally mediated control of the droplet formation in microchannels. The results show the possibility of using integrated microheater and temperature sensor for controlling droplet size during its formation. Generally, the droplet diameter is proportional to the ratio between the interfacial tension and the dynamic viscosity. In the low temperature range above room temperature, these parameters are both exponential functions of the temperature difference but of different exponents. As a result, droplet diameter can be controlled by the temperature of the breakup location. The method for controlling the droplet size reported here may play a role in designing microdispensers for droplet-based microfluidic systems. The experimental data may hint to the existence of different formation regimes in the characterized temperature range. If this hypothesis is correct, the device and experiments reported in this letter could be useful tools for the investigation of multiphase flow in microchannels. With only one set of liquids, several combinations of viscosity and interfacial tension can be realized in a single experiment by adjusting the temperature of the microheater.

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