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Thermocoalescence of microdroplets in a microfluidic chamber

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Droplet coalescence plays an important role in droplet-based microfluidics. This letter reports the phenomenon of thermocoalescence of two droplets in a chamber with a microheater. An integrated resistive sensor allows the measurement of heating temperature. The merging process was investigated at different flow rates. Experimental results showed that the droplet slows down at increasing temperature and eventually merges with the subsequent droplet. Coalescence occurs at a critical heating temperature. The letter discusses the relationship between droplet velocity, critical merging temperature, and flow rates. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4730606>]

Droplet-based microfluidics has been emerged as a promising platform for lab-on-a-chip (LOC) applications such as microreactor, protein crystallization,¹ and cell screening.² Advantages of this platform are the small sample volume, no cross-contamination, and efficient mixing.³ One of the basic operations of droplet-based microfluidics is the coalescence of droplets. However, merging of droplets with a size on the order of tens to hundreds micrometers is challenging due to the dominating surface tension. Droplet coalescence was induced by pillars embedded in a microchannel.⁴ Decompressing droplets also led to coalescence.⁵ Electrocoalescence has been reported as a common active approach to merge droplets. A pair of electrodes inside a microchannel can initiate localized droplet merging.^{6,7} Electrocoalescence requires precise alignment between electrodes and the microchannel, as well as a relatively high voltage. Temperature dependent viscosity and interfacial tension have been used for the manipulation of droplets.⁸ On an open and planar platform, droplets can be manipulated in a controlled manner using a micro heater array.^{9–11} Heat induced by microheaters was used to control the formation and switching process of droplets.¹² Another approach for inducing heat is employing a laser beam.¹³ Baroud *et al.*¹⁴ used a focused laser to manipulate, trap, and merge droplets. Temperature dependent droplet merging was demonstrated previously in a larger scale.¹⁵ However, a well controlled environment is needed to investigate the phenomenon of thermocoalescence of two droplets.

In this letter, we report the phenomenon of thermocoalescence of two droplets in a microchamber with integrated microheater and temperature sensor. Because of the small size, the required voltage is only a few volts, which is significantly lower than that of electrocoalescence. A resistive sensor is integrated into the device to provide a feedback on the heating temperature and hence precise control of the merging process. Water droplets in oil were formed at a T-junction upstream of the merging chamber. Fixing the flow rate of water, smaller droplets were generated with an increasing oil

flow rate. Expanding the channel at the merging location brings the droplets to a close contact. Increasing the applied voltage and consequently the heating temperature slowed down the droplets. Despite being in contact, coalescence only occurred if the temperature reached a critical T_{cr} . Following, the effect of the heating process on droplet velocities and the relationship between the critical temperature and the flow rate are investigated.

Figure 1 shows the schematic of the device used in our experiments. The microfluidic network was fabricated in polydimethylsiloxane (PDMS). Water droplets were formed at the T-junction and merged in a $1\text{ mm} \times 1\text{ mm}$ chamber. With the same height of $400\text{ }\mu\text{m}$, the widths of the microchannels for oil and water are $W_1 = 200\text{ }\mu\text{m}$ and $W_2 = 100\text{ }\mu\text{m}$, respectively. The microheater and the temperature sensor were fabricated on a Pyrex glass wafer by sputtering a Ti(50 nm)/Pt(500 nm) film and the subsequent lift-off process.

In our experiments, de-ionized (DI) water and mineral oil (Sigma M5904, 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 (2% by weight) to stabilize the formation of the water droplets. The liquids were delivered by two precision syringe pumps (KD Scientific, USA). The microheater was connected to a DC power supply (GW, Model GPC-30300). An epi-fluorescent inverted micro-scope (Nikon Eclipse TE 2000-E) and a high speed camera (Photron FASTCAM APX RS, USA) were used to capture the merging process at a

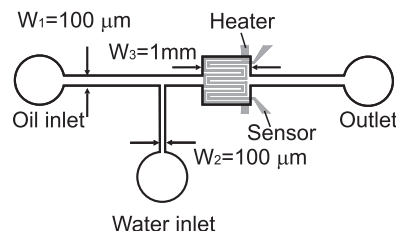


FIG. 1. Schematic of the microfluidic device for thermocoalescence. Water droplets are formed at a T-junction configuration. Droplets are merged in a heating chamber ($1\text{ mm} \times 1\text{ mm}$) where a microheater and microsensors are placed.

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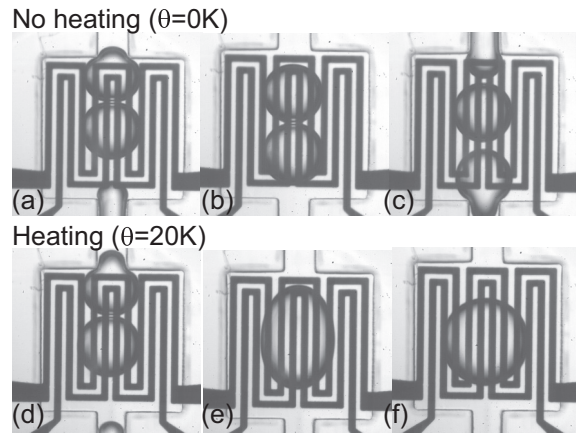


FIG. 2. Experimental results of thermocoalescence. (a)–(c) Two droplets in the merging chamber at room temperature. The droplets do not merge. (d)–(f) Two droplets in the merging chamber at a temperature of 45 °C. Merging occurs only at a temperature above the critical temperature, T_{cr} .

frame rate of 500 frames per second. The recorded images were then processed by a customized MATLAB program. The program converted the recorded grey-scale images into binary images and extracted the droplet velocity.

We kept the flow rate of water at a fixed value of $Q_w = 100 \mu\text{L/h}$ and increased the oil flow rate Q_o from 125 $\mu\text{L/h}$ to 225 $\mu\text{L/h}$. The applied voltage was calibrated to achieve a preset heating temperature. Figure 2 shows the typical behavior of two neighboring droplets in the chamber with and without heating at $Q_w = 100 \mu\text{L/h}$ and $Q_o = 200 \mu\text{L/h}$. In the chamber, the second droplet contacts the first droplet and forms a thin bridging film. If heat is not induced, this surfactant thin film is not broken¹⁶ and merging does not occur (Figs. 2(a)–2(c)). Both droplets are separated again while exiting the chamber. If a voltage is applied to the microheater, the first droplet slowed down due to thermocapillary force. The bridging film becomes thinner due to thermocapillarity. Rupture of the film and coalescence occurred only if the temperature reached a critical value of T_{cr} . We hypothesize that the bridging film becomes unstable at a high temperature. Film thinning at the rim of the contact area leads to subsequent ruptures.¹⁷ Figures 2(d)–2(f) show the typical thermocoalescence process of two water droplets at a temperature of 45 °C.

To investigate the influence of the temperature on the movement of a droplet, the velocity v at a given heating temperature T is normalized by its reference value at room temperature ($T_0 = 25 \text{ °C}$), $v^* = v(T)/v_0$. Since droplets were formed upstream of the merging chamber, the droplet size is independent of the heating temperature.

The temperature induced surface tension gradient leads to a thermocapillary force acting on the droplet and keeping it from moving forward. Gallaire *et al.*¹⁸ estimated this thermocapillary force as

$$F \simeq -\frac{\gamma' h L \mu_{oil}}{R(\mu_{oil} + \mu_{water})} \Delta T, \quad (1)$$

where γ' is the surface tension gradient, h is the channel height, L is the length of the chamber, R is the radius of the droplet, μ_{oil} is the viscosity of oil, μ_{water} is the viscosity of water, and ΔT is the difference between the chamber temper-

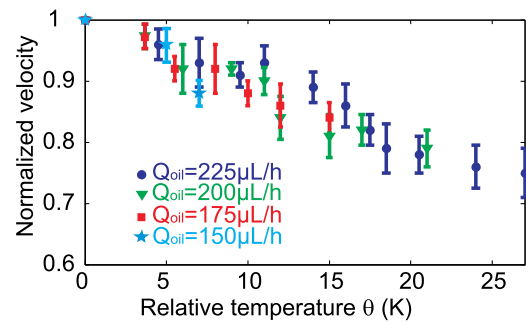


FIG. 3. Normalized droplet velocity versus relative heating temperature. Droplet velocities are normalized by their nominal values at room temperature.

ature and the upstream temperature. As the heating temperature increases, the thermocapillary force opposing the movement of the droplet also increases reducing its velocity. Figure 3 shows the relationship between the normalized droplet velocity and the relative heating temperature $\theta = T - T_0$. If the heating temperature is high enough, the droplets will be trapped inside the chamber. Beyond a critical value T_{cr} , coalescence of multiple droplets occurs.

Figure 4(a) shows that the critical temperature T_{cr} is linearly proportional to the flow rate of oil. We hypothesize that coalescence occurs because the surfactant thin film between two droplets become unstable. A merging temperature of T_m at the contact is required for coalescence. The temperature difference between the outlet and the inlet of the merging chamber is $T_{out} - T_{in} = 2\theta_m$, where $T_{in} = T_0$ and $\theta_m = T_m - T_0$ (Fig. 4(b)). Balancing the energy equation of forced convection for the merging chamber¹⁹

$$\dot{Q} \rho c 2\theta_m = h A \theta_{cr} \quad (2)$$

leads to the relationship between the critical heating temperature difference $\theta_{cr} = T_{cr} - T_0$ and the flow rate Q

$$\theta_{cr} = \frac{2\rho c \theta_m}{hA} Q, \quad (3)$$

where ρ , c , h , and A are the effective density, the specific heat, the heat transfer coefficient, and the surface area of the heater, respectively. This simplified model explains the linear

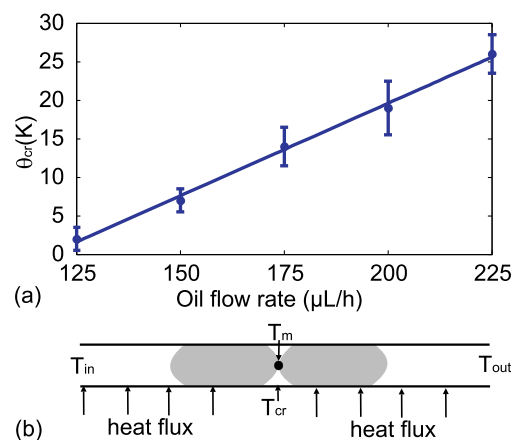


FIG. 4. (a) Critical temperature versus oil flow rate. (b) Cross-section view of the chamber with T_{in} , T_{out} , T_m are temperatures at chamber inlet, outlet, and middle of chamber. Contact point of droplets is at middle of chamber.

relationship between the critical temperature of the heater and the flow rate to maintain the required merging temperature T_m at the contact between the droplets. Thermocoalescence is more sensitive at a lower surfactant concentration. In another experiment where surfactant in the carrier fluid has a concentration of only 1% by weight, the value of the critical temperature is 5 °C lower than that shown in Fig. 4(a).

The present letter investigates the phenomenon of thermocoalescence in microscale using a microfluidic merging chamber with integrated microheater. The heating temperature is monitored by an integrated temperature sensor. Increasing the temperature leads to lower viscosity and surface tension and results in a higher thermocapillary force. The induced temperature field slows down droplets and eventually merges them at a critical value. The critical temperature is linearly proportional to the flow rate. Thermocoalescence is effective for merging droplets generated at a high flow rate ratio. Droplets are able to merge at a relatively low voltage as compared to electrocoalescence. The results reported here are useful for droplet merging and mixing in droplet-based lab-on-a-chip applications.

¹C. J. Gerds, V. Tereshko, M. K. Yadav, I. Dementieva, F. Collart, A. Joachimiak, R. C. Stevens, P. Kuhn, A. Kossiakoff, and R. F. Ismagilov, *Angew. Chem. – Int. Ed.* **45**, 8156 (2006).

- ²E. Brouzes, M. Medkova, N. Savenelli, D. Marran, M. Twardowski, J. B. Hutchison, J. M. Rothberg, D. R. Link, N. Perrimon, and M. L. Samuels, *Proc. Natl. Acad. Sci. U.S.A.* **106**, 14195 (2009).
- ³S. Y. Teh, R. Lin, L. H. Hung, and A. P. Lee, *Lab Chip* **8**, 198 (2008).
- ⁴X. Niu, S. Gulati, J. B. Edel, and A. J. Demello, *Lab Chip* **8**, 1837 (2008).
- ⁵N. Bremond, A. R. Thiam, and J. Bibette, *Phys. Rev. Lett.* **100**, 024501 (2008).
- ⁶C. Priest, S. Herminghaus, and R. Seemann, *Appl. Phys. Lett.* **89**, 134101 (2006).
- ⁷M. Zagnoni, C. N. Baroud, and J. M. Cooper, *Phys. Rev. E* **80**, 046303 (2009).
- ⁸N. T. Nguyen and X. Y. Huang, *Jpn. J. Appl. Phys.* **44**, 1139 (2005).
- ⁹Z. Jiao, X. Huang, N. T. Nguyen, and P. Abgrall, *Microfluid. Nanofluid.* **5**, 205 (2008).
- ¹⁰A. S. Basu and Y. B. Gianchandani, *J. Microelectromech. Syst.* **18**, 1163 (2009).
- ¹¹A. A. Darhuber, J. P. Valentino, S. M. Troian, and S. Wagner, *J. Microelectromech. Syst.* **12**, 873 (2003).
- ¹²Y. F. Yap, S. H. Tan, N. T. Nguyen, S. M. S. Murshed, T. N. Wong, and L. Yobas, *J. Phys. D: Appl. Phys.* **42**, 065503 (2009).
- ¹³K. T. Kotz, K. A. Noble, and G. W. Faris, *Appl. Phys. Lett.* **85**, 2658 (2004).
- ¹⁴C. N. Baroud, M. R. De Saint Vincent, and J. P. Delville, *Lab Chip* **7**, 1029 (2007).
- ¹⁵B. Xu, N. T. Nguyen, and T. N. Wong, *Biomechanics* **6**, 012811 (2012).
- ¹⁶J. Bibette, D. C. Morse, T. A. Witten, and D. A. Weitz, *Phys. Rev. Lett.* **69**, 2439 (1992).
- ¹⁷J. D. Chen, *J. Colloid Interface Sci.* **107**, 209 (1985).
- ¹⁸F. Gallaire, C. N. Baroud, and J. P. Delville, *Int. J. Heat Technol.* **26**, 161 (2008).
- ¹⁹N. T. Nguyen, D. Bochnia, R. Kiehnscherf, and W. Doetzel, *Sens. Actuators A: Phys.* **55**, 49 (1996).