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2013

Ho, P. C., & Nguyen, N.-T. (2013). Numerical study of thermocoalescence of microdroplets in a microfluidic chamber. *Physics of fluids*, 25(8), 082006-.

<https://hdl.handle.net/10356/101158>

<https://doi.org/10.1063/1.4819134>

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Citation: [Physics of Fluids \(1994-present\)](#) **25**, 082006 (2013); doi: 10.1063/1.4819134

View online: <http://dx.doi.org/10.1063/1.4819134>

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Numerical study of thermocoalescence of microdroplets in a microfluidic chamber

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(Received 7 December 2012; accepted 8 August 2013; published online 26 August 2013)

The present paper reports the numerical investigation of thermocoalescence of droplets in a microchannel network consisting of a droplet formation section connecting to a temperature-induced merging chamber. The numerical model is formulated as an incompressible immiscible two-phase flow problem with oil and water as the continuous and dispersed phase, respectively. The governing equations are solved using finite volume method on a staggered mesh. The interface is captured by a narrow-band particle level-set method. The paper examines the droplet formation process and droplet size at 4 different ratios of oil and water flow rate. The motion of the droplets from the formation section into and through the heat-induced merging chamber is analyzed. The numerical method is able to provide a visual presentation of the droplet movement in a heated environment under the influence of thermocapillarity. The relationship between the critical merging temperature and the fluid flow rate is also analyzed and discussed. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4819134>]

I. INTRODUCTION

In the last decade, the emergence of droplet-based microfluidics offers new functionalities for applications such as microreactor, on-chip polymerase chain reaction (PCR),¹ protein crystallization,² and cell screening.³ Small sample volume, efficient mixing, and low cross contamination are the key advantages of droplet-based microfluidics. The droplet acts as a carrier for chemical reagents or cells that can be transported and manipulated. Droplets are formed by co-flow, cross-flow, and flow-focusing configurations. Coalescence of individual droplets is required to facilitate mixing. Several methods have been developed to achieve droplet coalescence. Niu *et al.*⁴ explored the use of pillars in a microchannel to induce droplet coalescence. Tan *et al.*⁵ employed a trifurcating junction to redistribute equally spaced droplets bringing the droplets closer for coalescence. These methods are passive and geometrically determined, thus offer little control over the coalescence process. Electrocoalescence is an active method to achieve merging of droplet by electrically induce two droplets with opposite charges. Several electrocoalescence techniques have been reported in the literature.^{6–9} Most of these techniques require complicated electrode structures to be integrated in the microchannel adding complexity to device fabrication. Furthermore, a relatively high voltage is needed to allow coalescence to take place. Thermocoalescence offers an alternative solution for overcoming the drawbacks of passive methods and electrocoalescence. Thermocoalescence makes use of temperature dependent properties to achieve coalescence. The change in viscosity, surface tension, and the effect of surface tension gradient at the fluid interface (Marangoni effects) at elevated temperature are the mechanisms leading to coalescence. Luong *et al.*¹⁰ and Xu *et al.*¹¹ experimentally reported thermocoalescence of droplets in a heated merging chamber. The experiments showed a dependency of the critical temperature on the flow rate. However, much of the physics governing thermocoalescence remains unclear. Due to the complex geometry and the strong coupling of the different physical fields, further understanding requires a numerical investigation of

the phenomenon. Computational requirement for simulating a three-dimensional droplet formation process is daunting. To the best of our knowledge, no numerical experiment on thermocoalescence has been reported in the literature. The present paper reports the use of Level Set Method (LSM) to investigate thermocoalescence in a heated microchamber. The present work discusses the physical phenomena leading to the reduction of the gap between two adjacent droplets. A condition is reached where two droplets are in contact with each other leading to subsequent coalescence. However, the present numerical model does not cover the molecular level of interaction between the droplets, especially in the bridging film between the two droplets. Instead, the work examines in detail the time history of the velocity of the droplets in the merging chamber. Further to previous experimental heater configuration, another configuration is also considered in the present study to understand its impact on the critical merging temperature. LSM has been used in our previous works to study the formation of droplet at a thermally controlled T-junction.¹² LSM proves to be a powerful tool for solving complex deformation and for reconstruction of implicit surfaces in a fixed rectangular grid.¹³ LSM is able to handle challenging problem such as coalescence and breakup of droplets. The paper starts with a brief description of the problem, its mathematical formulation, solution procedure, followed by the discussion of the results and the concluding remarks.

II. NUMERICAL MODELING

Figure 1 shows the geometry of the microchannel of the present study. The microchannel system is similar to that used in the experiments of Luong *et al.*¹⁰ The network consists of a T-junction and a 1-mm $\times$ 1-mm merging chamber. The T-junction has a square cross section and two inlets — one for the continuous phase (oil) and the other for the dispersed phase (water). The continuous phase and the dispersed phase are immiscible. For the convenient of the later implementation of level-set simulation, the phases are designated as the negative fluid (–ve) and the positive fluid (+ve) for the continuous and dispersed phase, respectively. These phases are distinguished in the level set function by their signs. The continuous phase (–ve fluid) flows into the channel from the left inlet with an average inlet velocity of u_{-o} and an initial temperature of T_o . The dispersed phase (+ve fluid) enters the other inlet with an average velocity of u_{+o} at T_o . Droplet formation occurs at the T-junction where the two channels join. The shear force and the pressure buildup at the junction causes a breakup of the disperse phase and as a result, droplets of the +ve fluid are formed. A layer of oil always exists between the droplet and the channel wall. Therefore, the droplet will not come into direct contact with the wall. Thus, there is no moving three-phase contact line. Based on this assumption, the contact angle does not need be taken into account in our problem. To implement this assumption, the present numerical model assigned a thin layer of 10 μm thickness adjacent to the

FIG. 1. Thermally controlled droplet coalescence in a microchannel with –ve fluid (oil) as continuous phase and +ve fluid (water) as dispersed phase.

channel wall to be filled only by continuous fluid. The droplets flow downstream into the merging chamber where droplet coalescence occurs under a thermally controlled condition. Fully developed flows are assumed for both inlets. Non-slip condition is enforced at the walls of the channel. Outflow boundary condition is assigned for the outlet.

The temperature field is induced into the merging chamber by a heater attached to the wall as shown in Figure 1. The boundary conditions are set such that the temperatures of the locations on the wall directly adjacent to the heater are set to T_{set} . Other locations of the wall which are outside the heater are maintained at T_0 . At the outlet, a zero temperature gradient condition is employed. With these boundary conditions, a non-uniform temperature field is established in the channel and changes the properties of both fluids. This temperature field induces additional forces acting on the droplet, and together with the changes of fluid properties in the merging chamber giving the right condition for coalescence.

A. Mathematical formulation

Level-set method¹⁴ is used to capture the interface between the two immiscible phases. The domain of interest consists of the $-ve$ fluid and the $+ve$ fluid. These fluids are separated by the interface Γ . A level-set function, ϕ , is used to describe the distance from the interface and can be expressed mathematically as

$$\phi = \begin{cases} -d, & \vec{x} \in \Omega_- \\ 0, & \vec{x} \in \Gamma \\ +d, & \vec{x} \in \Omega_+ \end{cases}, \quad (1)$$

where d is the shortest normal distance from the interface Γ , for any location, $\vec{x}$. Heaviside function is used to handle the combination of fluid property near the interface. Any property α of the special fluid can be conveniently expressed using the Heaviside function H :

$$\alpha(\phi) = (1 - H)\alpha_- + H\alpha_+, \quad (2)$$

where H is

$$H(\phi) = \begin{cases} 0, & \phi < -\varepsilon \\ \frac{\phi + \varepsilon}{2\varepsilon} + \frac{1}{2\pi} \sin\left(\frac{\pi\phi}{\varepsilon}\right), & |\phi| \leq \varepsilon \\ 1, & \phi > +\varepsilon \end{cases} \quad (3)$$

with H smears over a thickness of 2ε and ε is set to be 1.5 times of the grid size.

The evolution of the fluid interface Γ on a rectangular grid with a grid size of Δx is governed by

$$\phi_t + \vec{u} \cdot \nabla \phi = 0. \quad (4)$$

To maintain ϕ as a signed distance function, a reinitialisation procedure is used. ϕ is set to the steady-state solution of the following equation:

$$\frac{\partial \phi'}{\partial \bar{t}} = \overline{\text{sign}}(\phi') (1 - |\nabla \phi'|), \quad (5)$$

where $\bar{t}$ is a pseudo time for a second level-set function that is employed for redistancing purpose only with the following initial condition:

$$\phi'(\vec{x}, 0) = \phi(\vec{x}). \quad (6)$$

From Eq. (5), the steady state solution satisfies $|\nabla \phi'| = 1$. The initial condition in Eq. (6) ensures that the interface value, ϕ' , is the same as ϕ . Therefore, the steady state values of ϕ' are the distances from the interface.

The conservation of mass, momentum, and energy of the special fluid are used to describe the two-phase microfluidic system and are formulated mathematically as

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{u}) = 0, \quad (7)$$

$$\frac{\partial (\rho \vec{u})}{\partial t} + \nabla \cdot (\rho \vec{u} \vec{u}) = -\nabla p + \nabla \cdot [\mu (\nabla \vec{u} + \nabla \vec{u}^T)] + \vec{f}_F, \quad (8)$$

$$\frac{\partial (\rho c_p T)}{\partial t} + \nabla \cdot (\rho c_p \vec{u} T) = \nabla \cdot (k \nabla T), \quad (9)$$

where ρ , μ , c , and k are the density, the dynamic viscosity, the specific heat, and the thermal conductivity, respectively. In Eq. (9), the interfacial force is given as¹⁵

$$\vec{f}_F = -\kappa \sigma \hat{N}_F D(\vec{x} - \vec{x}_F) + (\hat{N}_F \times \nabla \sigma) \times \hat{N}_F D(\vec{x} - \vec{x}_F), \quad (10)$$

where the Dirac delta function $D(\vec{x} - \vec{x}_F)$, the unit normal to the interface $\hat{N}_F$, and the curvature κ are defined respectively as

$$D(\vec{x} - \vec{x}_F) = D(\phi) = \begin{cases} \frac{1 + \cos(\pi(\phi)/\varepsilon)}{2\varepsilon} & |(\phi)| < \varepsilon \\ 0 & \text{otherwise} \end{cases}, \quad (11)$$

$$\hat{N}_F = \frac{\nabla(\phi)}{|\nabla(\phi)|},$$

$$\kappa = \nabla \cdot \hat{N}_F.$$

The first and second terms in (10) account for capillarity and thermocapillarity effects, respectively.

The conservation Eqs. (7)–(9) are solved on a staggered grid system by employing the Semi-Implicit-Method for Pressure Linked Equation Revised (SIMPLER) algorithm¹⁶ to resolve the pressure-velocity coupling. In the SIMPLER algorithm, the discretised momentum equation will have the form¹⁶

$$a_{i,j} \vec{u}_{i,j}^* = \sum a_{nb} \vec{u}_{nb}^* + p_{i,j}^* A_{i,j} + b_{i,j}, \quad (12)$$

where coefficients, $a_{i,j}$ and a_{nb} contain a combination of advective flux and diffusive conductance at the control volume cell face, $A_{i,j}$ is the cell face area of the control volume and $b_{i,j}$ is the momentum source term which contain the interfacial force.

The equation is solved using the guessed pressure field, p^* to yield the velocity components $\vec{u}^*$. Next, the velocity components are used to solve pressure correction term. The guessed pressure and velocity component are then corrected. Finally, the new pressure is set as the next guessed pressure and the procedure is repeated. Details of the procedure is explained in Ref. 16. The iteration will continue until the source, b , becomes sufficiently small everywhere in the discretised domain. The source term, b , is the corrective factor of the guessed pressure for the each subsequent iteration. For convergence, the value of mass source term will practically become zero for all the control volumes. The criteria for convergence are as follows:

- (1) The maximum value of the source term for all the control volume is less than 1×10^{-12} ; and
- (2) The summation of source term for all the control volume is less than 1.

The evolution of the level-set function and its redistancing procedure are implemented in a narrow-band procedure.¹⁷ For redistancing, the stopping criterion employed by Sussman and Fatemi¹⁸ is adopted. As mentioned earlier, the interface thickness is ± 1.5 times the grid size, Δx . Thus, we perform re-initialization up to $\bar{r} = 3\Delta x$. Both level-set and redistancing functions are spatially discretized with Weighted Essentially Non-oscillatory 5th order (WENO5) algorithm¹⁹ and advected using Total Variation Diminishing Runge-Kutta 2nd order (TVD-RK2) algorithm.²⁰ For the redistancing procedure, the value of the pseudo time step, $\Delta \bar{r}$, is $1/4\Delta x$. It has been reported

in Ref. 18 that WENO5 scheme has a gain in accuracy which has helped to reduce long iteration time. Internally in our simulation program, we use a mass loss index to monitor the health status of our numerical solution. The requirement is not more than 10% mass loss. The mass loss index is described as below:

$$\Delta M = \frac{M_d - M_c}{M_d}, \quad (13)$$

where M_d and M_c are the desired mass and the most current mass of the reference phase, respectively. The values are computed purely base on the volume flow rate of fluid and the calculated mass of the fluid from the numerical results of each iterations cycle.

The overall solution procedure is as follow. Given known f , $\vec{u}$, p , and T , proceed the solution in time for f^{n+1} , $\vec{u}^{n+1}$, p^{n+1} , and T^{n+1} :

- Step (1) Set $\vec{u}^{n+1} = \vec{u}^n$;
- Step (2) Compute ϕ^{n+1} from Eq. (4);
- Step (3) Perform redistancing via Eqs. (5) and (6);
- Step (4) Calculate the properties of the special fluid using Eqs. (2) and (3);
- Step (5) Solve for $\vec{u}^{n+1}$ and p^{n+1} from Eqs. (7) and (8) via the SIMPLER algorithm;
- Step (6) Solve for T^{n+1} from Eq. (9);
- Step (7) Iterate step (2) through (6) and check if solution converges. Solution is said to converge when the correction factor of the SIMPLER algorithm is less than 1×10^{-12} after a minimum number of 10 iterations;
- Step (8) Proceed to the next time step by starting from step (1) if solution converges. Otherwise, terminate the procedure.

B. Numerical setup

As in the experiments, the flow rate of water is kept at $Q_+ = 100 \mu\text{l/h}$ in combination with the various flow rate for oil at $Q_- = 150 \mu\text{l/h}$, $175 \mu\text{l/h}$, $200 \mu\text{l/h}$, and $225 \mu\text{l/h}$.

The viscosity μ and the surface tension σ depend on temperature. These temperature dependent properties normalized by their corresponding values at $T_o = 25^\circ\text{C}$ were reported previously by our group in Ref. 21 and as shown below:

$$\sigma^* = e^{-0.0144\Delta T}, \quad (14a)$$

$$\eta^* = e^{-0.0344\Delta T}, \quad (14b)$$

where $\sigma^* = \frac{\sigma}{\sigma_o}$, $\eta^* = \frac{\eta}{\eta_o}$, and $\Delta T = T - T_o$. σ_o and μ_o are the surface tension and viscosity at reference temperature, $T_o = 25^\circ\text{C}$.

The densities, viscosities, and interfacial tension of the fluids at reference temperature of $T_o = 25^\circ\text{C}$ are $\rho_- = \rho_+ = 1000 \text{ kg/m}^3$, $\mu_{-o} = 2.64 \times 10^{-2} \text{ Pa s}$, $\mu_{+o} = 1.00 \times 10^{-3} \text{ Pa s}$, and $\sigma_o = 3.65 \times 10^{-3} \text{ N/m}$. The densities of the fluids are set to be the same since the densities of both fluids are considered as temperature independent and buoyancy is thus neglected.²¹⁻²³

Table I summarizes the respective Reynolds number and capillary numbers for the various combinations of flow rates.

TABLE I. Re and Ca for different combinations of flow rates.

Q_- flow rate ($\mu\text{l/h}$)	Q_+ flow rate ($\mu\text{l/h}$)	Re	Ca
150	100	0.395×10^{-2}	0.753×10^{-2}
175	100	0.460×10^{-2}	0.879×10^{-2}
200	100	0.526×10^{-2}	1.005×10^{-2}
225	100	0.592×10^{-2}	1.130×10^{-2}

FIG. 2. Mass loss for different grid size.

Although it is not described in the present study, the optimization of LSM settings was carried out as per our previous work.¹² It is based on the specific set of hydrodynamic condition used in the experiment. A pre-check simulation to determine a computationally economical grid size is done using the geometry of the forming section of the microchannel (inset of Figure 2). Grid size of $\Delta x = 14 \mu\text{m}$ and $10 \mu\text{m}$ are tested.

As shown in Figure 2, the grid size of $\Delta x = 14 \mu\text{m}$ did not yield accurate result as compared to that of $\Delta x = 10 \mu\text{m}$ for a time step of 1.00×10^{-4} s. The mass loss for $\Delta x = 14 \mu\text{m}$ is much higher. Therefore, a grid size, Δx , and time step of $10 \mu\text{m}$ and 1.00×10^{-4} s, respectively are employed for this present study.

III. RESULTS AND DISCUSSIONS

In this section, the droplet formation process and droplet size at various liquid flow rates are discussed. The motions of the droplet from the droplet formation section to the merging chamber are examined. Finally, the relationship of the merging temperature and flow rates is discussed.

In the present numerical study, coalescence of the water droplets at various flow rate ratios and temperatures of the merging chamber are considered. The numerical results are compared with the experimental data reported previously by our group.¹⁰ At the upstream of the merging chamber, the formation of water droplet is realized by a T-junction as shown in Figure 1. The size of the droplets and their rate of formation are determined by the flow rate ratio between the continuous phase (oil) and the dispersed phase (water).

A. Droplet formation

This section discusses the formation of droplet at the T-junction of the microchannel device under the flow conditions employed in our previous experiments.¹⁰

During the droplet formation process, the forces acting on the forming droplet are the pressure force F_p , the viscous force F_v , and the interfacial tension force F_σ as depicted in Figure 3(b). F_p is caused by the streamwise pressure difference across the T-junction section and increases when the resistance to flow inside the microchannel increases. F_v is caused by the viscous stress of the continuous phase acting on the forming droplet. This force is proportional to the viscosity and the

FIG. 3. Droplet formation process: (a) experiment¹⁰ and (b) present numerical procedure.

velocity gradient. F_σ withholds the forming droplet from detachment and is proportional to the interfacial tension σ and the circumference of the water thread l .

Figure 3 compares the formation of a droplet from the simulation and the experiment. It can be noticed that the shape of the droplet tip and the shape of droplet at breakup are slightly different for the numerical results. This may be attributed to the presence of surfactant which lowers the surface tension and makes the droplet easier to break up. However, the physics of droplet formation process remains consistent.

When water enters the microchannel, the interface between the two fluids is formed, Figure 3(b). As the dispersed phase grows, it progressively blocks the cross section of the main channel resulting in an increase of F_p due to the increase in flow resistance. A region of high pressure builds up as indicated in Figure 3. At the same time, F_v increases due to the increase in surface stress on the interface. Both F_p and F_v continue to act on the growing droplet, pushing it downstream, stretching the water thread and thinning its circumference. As the circumference of the water thread reduces, the interfacial tension squeezes the water within the neck of the developing droplet away from the neck and further thinning it. This process continues until a point when F_σ is no longer able to hold the droplet. The water thread breaks and the droplet is formed. This is a case of droplet formation in the squeezing regime where the pressure force F_p and viscous force F_v play the critical role in the droplet breakup. De Menech *et al.*²² defined the different regimes of droplet formation based on the range of capillary number. Squeezing regime is observed with a capillary number Ca approximately less than 0.01. Dripping regime happens with Ca above 0.02. In the present study, the capillary number ranges between 0.008 and 0.011 and squeezing regime can be observed from the numerical results.

Figure 4 shows that the size of the droplet decreases with increasing flow rate. Higher oil flow rate leads to higher pressure force and viscous force to overcome the interfacial force. As result, a shorter time is needed to achieve droplet breakup at higher flow rate and the droplet size reduces

FIG. 4. Simulated droplet volume at different oil flow rates.

almost linearly. A similar trend was also observed experimentally by Garstecki²³ where the droplet length, or indirectly the droplet volume, reduces with increasing flow rate ratio.

B. Movement of droplet in the merging chamber at elevated temperature

Figure 1 shows the heater attached to the wall of the merging chamber. The temperature of the wall adjacent to the heater is raised to T_{set} when the heater is switched on. Figure 5(a) shows the temperature profile in the merging chamber when the heater is set to 60°C . Because the heater is attached to one side of the merging chamber, the temperature distribution within the chamber is asymmetrical. Figure 5(a) also shows that the droplet is skewed to one side as it enters the heated merging chamber. The asymmetrical temperature field draws the droplet towards the region

FIG. 5. (a) Temperature profile and position of droplet in the heated merging chamber at 60°C ; (b) droplet position in the merging chamber at 25°C and 60°C ; and (c) droplet position in the merging chamber at 60°C with extended outlet length.

of higher temperature close to the heated side of the chamber creating a gap between the droplet and the opposite side of the chamber wall.

The gap has a significant impact on the droplet movement. It allows oil to bypass the droplet instead of pushing directly on the droplet. Under this condition, the hydrodynamic drag force on the droplet is reduced resulting in a decrease in velocity of the droplet in the forward direction as it travels across the chamber. This phenomenon can be analyzed by comparing the movement of the droplet in the merging chamber in the cases of 25°C and 60°C at an oil flow rate of 225 $\mu\text{l/h}$ and a water flow rate of 100 $\mu\text{l/h}$, Figure 5(b). By the time the droplet at 25°C already exits (0.68 s), the droplet at 60°C chamber temperature still remains in the chamber. These results show that the droplet velocity decreases at elevated temperature.

To minimize computational resources, the channel outlet length is only kept to 100 μm . However, a simulation is carried out with an extended outlet length of 200 μm to understand if there is any sensitivity of the outflow condition has on the numerical results. As shown in Figure 5(c), the outlet length has no significant impact to the results. Thus, we maintain 100 μm outlet length for the subsequent numerical experiments carried out in the present paper.

Luong *et al.*¹⁰ reported in their experiment that the droplet velocity decreases with increasing heating temperature, Figure 6(a). The droplet velocity is normalized by values at $T_o = 25^\circ\text{C}$ such that $v^* = v/v_o$. The experimental data showed that the normalized droplet velocity is exponentially proportional to the heating temperature as

$$v_w^* \propto \frac{\sigma^*}{\eta_w^*} \propto \exp\left(-\frac{\theta}{\theta_o}\right), \quad (15)$$

where $\theta = T - T_o$.

From the present numerical data for a flow rate ratio of 150/100, the simulated velocity also shows an exponential decay over increasing temperature, Figure 6(b).

Next, we examine the time history of the velocity. Figure 7(a) shows the velocity of the 1st and 2nd droplet in the merging chamber for the case of 25°C, 50°C, and 60°C based on the present numerical procedure. The velocity of the droplet is normalised to its initial velocity in the

FIG. 6. Relationship between droplet velocity and increase in temperature: (a) experiment and (b) present numerical procedure.

FIG. 7. Velocity of droplet across the merging chamber: (a) present numerical procedure and (b) experiment with 60°C.

forming section of the microchannel device before entering the chamber. The three cases represent the following scenarios: (i) 25°C, no heating; (ii) 50°C, heating but no droplet coalescence; and (iii) 60°C, heating and droplet coalescence.

Figure 7(b) shows the corresponding velocity profile of the droplet taken from the actual experiment. The present numerical solution and experiment data show similar trends. For all three cases, the present numerical solution shows that the velocity of the first droplet decreases as it enters the chamber. The expansion in the channel geometry induces a sudden decrease in flow velocity leading to a slowdown in the droplet velocity. This behaviour can also be observed in the experiment, Figure 7(b).

At elevated temperatures (i.e., 50°C and 60°C), the droplet accelerates upon entry into the chamber. Such behaviour cannot be observed in the case of no heating (i.e., 25°C) and is attributed to the thermocapillary effect.

Figure 8(a) shows the shape of droplet superimposed on the temperature distribution within the chamber. The droplet is more spherical without heating. With heating, different temperatures are experienced by the leading and the trailing edges of the droplet. The trailing edge is colder than the leading edge resulting in a higher surface tension at the trailing edge. The surface tension gradient experienced by the droplet affects the otherwise spherical shape. Due to the higher surface tension, the radius of curvature of the trailing face is higher than the leading face. The difference in surface tension accelerates the droplet forward. The rate of acceleration increases with the chamber temperature. As shown in Figure 8(b), the normalized velocity of the droplet reaches 1.19 for 60°C at the instant when the droplet is completely inside the chamber. Whereas, the value at 50°C is only 1.02. However, as the droplet transverses further into the chamber, its velocity reduces. This result

FIG. 8. (a) Droplet shape at position right after entry point of merging chamber and (b) velocity of droplet versus chamber downstream distance.

demonstrates the effect of thermocapillary force at elevated temperatures. The effect is not so obvious in the experiment although a small “bump” in the velocity profile can be seen at the beginning of the drop movement in the heated chamber. Further investigation is needed to understand if the presence of surfactant is preventing this effect. However, this is beyond the scope of the present work.

The thermocapillary effect begins to wear off quickly as the droplet travels further into the chamber. A progressive reduction in droplet velocity can be observed until the droplet reaches the chamber exit. With no heating, the velocity of the droplet does not change significantly after the initial decrease in velocity. This trend is in good agreement with the experimental data.

A further investigation was conducted to understand the magnitude of influence of the thermocapillary force as compared to the hydrodynamic drag force of the oil on the droplet movement. Two simulations were carried out by limiting the change in fluid viscosity and surface tension with temperature. The separation of temperature dependencies of viscosity and interfacial tension is not possible in the real situation and such investigation can only be carried out numerically instead of the actual experiment. As shown in Figure 9, the velocity of the droplet at elevated temperature is almost the same as at 25°C if the surface tension is kept constant, e.g., temperature independent. This result indicates the significance of thermocapillary force on the thermocoalescence process. On the other hand, the velocity profile of the droplet is close to the case of elevated temperature if the viscosity is kept constant. This result further ascertains the dominating effect of thermocapillary force on the droplet movement.

As mentioned in Eq. (3), the thickness of fluid interface for the present study is described by smearing factor of 1.5 times of grid size, Δx . A smearing factor of 3 times Δx is employed in a separate simulation to check the sensitivity of smearing factor on the numerical results. It has been observed in Figure 9 that the droplet movement is the same as the reference and coalescence also happened at 60°C. The results have shown that changing the smearing factor does not significantly affect the numerical results.

FIG. 9. Velocity of the droplet over time for viscosity only and interfacial tension only as temperature dependent parameters as well as for an increased smearing factor.

C. Droplet coalescence

The mechanism behind droplet coalescence is the rupture of the bridging oil film between two droplets due to their close proximity. The high surface curvature at the liquid bridge results in high capillary pressure around the bridge pulling the two droplets together, leading to coalescence.^{24,25} The idea of having a merging chamber is to slow down the droplet and to close up the distance between two adjacent droplets. To enhance the condition of droplet coalescence, heat is induced to further slowdown the first droplet waiting for the next droplet. As explained in Sec. III B, increasing the temperature of the merging chamber will lower the viscosity of the fluid as well as creating an imbalance in surface tension of the droplet and reducing the velocity of the droplet. Thus, our present simulation cannot model the microscopic coalescence process. Coalescence occurs numerically as the oil bridge between the droplets is less than the grid size.

Figure 10 shows that droplet coalescence occurs at 60°C at an oil-to-water flow rate ratio of 225/100. With heating, the droplet slows down and arrives near the exit only at 680 ms as compared to 560 ms with no heating. The droplet in a non-heated chamber exits the channel much earlier at 640 ms, Figure 10(a). However, the droplet in the heated chamber can be observed to remain at almost the same position near the exit after 680 ms, Figure 10(b). When the droplet approaches the exit of the chamber, its leading edge experiences a lower temperature than the trailing face. The colder leading face has a higher surface tension. The thermocapillary force pushes the droplet backward acting against the forward hydrodynamic drag of the fluid flow. When force equilibrium is achieved between these two forces, the droplet appears to be “trapped” at the exit of the merging chamber waiting for the next droplet to arrive. The incoming droplet eventually comes into contact with the trapped droplet resulting in coalescence.

After coalescence, the merged droplet is twice as big and blocks the exit of the merging chamber, increasing its pressure. The pressure builds up in the merging chamber until it can push the merged droplet out of the chamber.

In the present work, the key factor for droplet coalescence depends on the ability to trap the droplet at the exit of the merging chamber. Coalescence relies on the thermocapillary force to counter the hydrodynamic force of the fluid flow. Both thermocapillary and hydrodynamic force are temperature dependent. However, hydrodynamic force is also dependent on fluid flow rate. For a particular flow rate, there is a critical temperature beyond which droplet coalescence will take place.

FIG. 10. Droplet coalescence at elevated temperature: (a) no heating and (b) heating at 60°C.

We describe the difference between this threshold temperature and room temperature of 25°C as the critical merging temperature, θ_{cr} :

$$\theta_{cr} = T_{merging} - T_o.$$

As depicted in Figure 11(a), the present numerical study shows that the critical temperature increases with increasing flow rate. In order to trap the droplet in the chamber for coalescence to happen, a higher thermocapillary force is required to overcome the hydrodynamic force of a higher flow rate. Thus, a higher temperature is needed to achieve the force equilibrium. This trend agrees with our previous experiments.¹⁰ However, the critical merging temperature for the range of oil flow rate from 27°C to 30°C for the present numerical study which is much smaller than that of the experiment. This difference may be caused by the dynamic accumulation of surfactant molecules at the interface which is not considered in the present numerical model. Lu and Corvalan²⁶ discussed the effect of surfactant on droplet coalescence.

Our numerical study was further extended to a merging chamber with a front heater in addition to the original back heater. The additional heater allows a wider span of the critical merging temperature as shown in Figure 11(a).

In the case with only a back heater, the asymmetric temperature distribution in the merging chamber causes the droplet to be drawn towards the heated side resulting in a gap between the droplet and the wall, Figure 5(a). The fluid is allowed to bypass the droplet, and the droplet experiences a lower hydrodynamic drag. However, in the case of two heaters, the symmetric temperature distribution within the chamber keeps the droplet in the middle of the chamber and occupies almost the entire cross section of the channel with no gap between the droplet and the channel wall, Figure 12. Thus, the droplet fully experiences the hydrodynamic force from the fluid flow. In order to counteract this hydrodynamic force so that the droplet can be “trapped” to meet the incoming second droplet, a higher thermocapillary force is necessary. This force can only be achieved with a higher temperature. Thus a higher critical merging temperature is obvious at a higher oil flow rate.

The effect of hydrodynamic and thermocapillary force is even more evident by comparing the velocity of the droplet and the average velocity of the surrounding oil for the two different heater

FIG. 11. Critical merging temperature versus flow rate: (a) present numerical results and (b) experiment.

configurations as shown in Figure 13. Unlike the case of one heater, no initial acceleration of the droplet at the point of entry into the merging chamber can be observed for the case of two heaters. The thermocapillary force is smaller. This is due to a more evenly distributed surface tension gradient on the droplet as a result of the symmetrical temperature field. It can also be observed in Figure 13(b) that the oil velocity is lower than the case of no heating. Coupled with a decrease in viscosity of oil

FIG. 12. Temperature profile with heaters on both sides of the chamber.

FIG. 13. (a) Velocity of droplet across the merging chamber for different heater configurations and (b) oil velocity for different heater configurations.

at elevated temperature, the hydrodynamic drag force on the droplet is expected to be lower than the no-heating case. However, the droplet velocity can still maintain quite close to that of no-heating case because of the smaller thermocapillary force.

In general, the oil velocity correlates with the droplet velocity. The oil velocity is more oscillatory for the case of one heater. As explained earlier, the droplet is drawn to one side of the wall and some oil can bypass the droplet through the gap between the droplet and the opposite side of the wall. This could induce some disturbance to the hydrodynamic force equilibrium surrounding the droplet and thus leading to the unstable oil velocity.

Further down the chamber, the droplet velocity for both heater configurations continue to reduce as compared to the case of no heating. For the case of one heater, it can be understood that the gap for oil to bypass has significantly reduced the effect of hydrodynamic force. As a result, the one heater case has the biggest reduction in droplet velocity among the 3 cases.

IV. CONCLUSIONS

The present paper reports a three-dimensional numerical simulation of thermocoalescence in a heated chamber with oil as the continuous phase and water as the disperse phase. The flow rate of

oil ranges between 150 $\mu\text{l/h}$ to 225 $\mu\text{l/h}$ with a constant water flow rate of 100 $\mu\text{l/h}$. The process of droplet formation is validated with the experiment data and shows a good agreement.

Based on the numerical results, the size of the formed droplet reduces with increasing oil flow rate. The higher viscous force from the increased oil flow rate causes breakup to occur earlier leading to a small droplet and a higher formation frequency. The movement of droplet in the heated chamber was examined. The numerical results show that the velocity of the droplet is reduced across a heated chamber. The droplet initially accelerates and thereafter slows down as it travels through the chamber. When subjected to the temperature gradient of the heated chamber, the temperature dependent surface tension between the leading and trailing edge of the droplet pushed the droplet towards the heated region. Analysis on the adjacent droplet shows similar trend of the velocity as compared to the first droplet although the normalized velocity is slightly lower. At a critical temperature, the droplet remains at the same position near the exit of the merging chamber. This phenomenon favors coalescence by holding the droplet at the chamber exit while waiting for an incoming droplet. Incidentally, the critical temperature for droplet coalescence is dependent on the flow rate. The numerical results show that at a higher oil flow rate, a higher temperature is required for droplet coalescence. The relationship between the critical temperature and the flow rate is almost linear. A similar trend can be seen in the experiment except that the temperature range is wider for the same range of oil flow rate. The numerical experiment of droplet coalescence was further extended to a configuration with front and back heaters. The configuration with one heater requires a lower critical merging temperature than the configuration with two heaters.

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