Sample Transport with Thermocapillary Force for Microfluidics

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Abstract. This paper presents a novel concept for transport of aqueous sample in capillaries. The concept is based on the thermocapillary effect, which utilizes the temperature dependency of surface tension to drive a sample droplet. To date, the major problem of this concept was the evaporation of the aqueous sample. In our approach, a liquid-liquid system was used for delivering the sample. The aqueous sample is protected by silicone oil, thus evaporation can be avoided. A transient temperature field drives both liquids away from a heater. The paper first presents a theoretical model for the coupled thermocapillary problem. Next, the paper compares and discusses experimental results with different capillary sizes. The results show the huge potential of this concept for handling sample droplets dispersed in oil, which are often created by droplet-based microfluidics.

Keywords: micro plug, micro droplet, thermocapillary, digital microfluidics, multi-phase flow, MEMS.

1. Introduction

Recently, a shift from continuous-flow system to droplet-based system has been observed in microfluidic research. Microdroplets formed in microchannels were used as a mean for reagent transport and a platform for chemical reactions. A micro droplets are called a micro plugs if its length is larger than the channel width. The tension at these solid/liquid/liquid or solid/liquid/gas interfaces can be manipulated by electrostatic force or temperature. Pollack et al. [1] used electrowetting effect to manipulate surface tension across an aqueous droplet suspended in oil and subsequently to transport a droplet. Ren et al. presented a theory for droplet transport based on electrowetting [2]. Brozoska et al. [3] investigated the movement of a droplet on a flat surface, which is exposed to a temperature field. Darhuber et al. [4] and Tseng et al. [5] recently studied the dynamic behaviors of microdroplets exposed to the temperature gradient of a flat plate. The heat transfer inside a droplet was analyzed by Sammarco and Burns[6]. Yarin et al. [7] studied the transient behavior of droplets surrounding a thin fiber with a temperature jump as the initial condition. Nguyen and Huang reported a one-dimensional
(1-D) analytical model for the transient behavior of a single-phase liquid droplet in a capillary with a jump of heat flux as the initial condition [8]. Glockner and Naterer later investigated the same problem with a two-dimensional (2-D) numerical model and microchannels fabricated in silicon [9].

Micro droplets dispersed in oil become increasingly popular with a number of interesting application such as chemical analysis and protein crystallization [10, 11]. In these applications, microdriplets are often generated in a microfluidic device and stored as liquid plugs in a glass capillary for storage or further analysis [12]. Chan and Yang reported a two-liquid delivery concept based on passive capillary effect [13]. To the best of our knowledge, no work has been reported on the active actuation of micro plugs stored in capillaries. This paper reports a method for transporting aqueous sample plug in a glass capillary. We consider the simplest system consisting of a oil plug and a water plug. This liquid/liquid system is then driven by thermocapillary forces, which are induced by the transient temperature field of the capillary. A heater at one end of the capillary controls this temperature field and consequently drives the liquid plugs. We first show the theoretical model of this concept. Next, the experimental setup and materials are described. Droplet positions are then compared and discussed according to varying parameters such as capillary size and liquid system.

2. Theory of sample transport based on thermocapillary effect

Figure 1 depicts the model for the actuation scheme described in this paper. If heat radiation and thermal coupling between the capillary wall and the plugs are negligible, heat transfer in the capillary wall is governed by the one-dimensional transient equation:

\[
\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} - \frac{2h}{\rho c R_o} T.
\]  

where \( T \) is the temperature relative to the ambient temperature, \( R_o, \alpha, \rho, c \) are the outer radius, the thermal diffusivity, density and specific heat capacity of the capillary, respectively. The initial boundary conditions for eq. (1) are:

\[
t = 0 : \quad T(x) = 0
\]

\[
t > 0 : \begin{cases}
  x = 0, & \frac{dT}{dx} = -\frac{q''}{k} \\
  x = L_c, & T = 0
\end{cases}
\]

where \( L_c, q'', k \) are the length of the capillary, the heat flux and the heat conductivity of the capillary, respectively. Introducing dimension less variables \( T^* = T(q'' L_c/k), x^* = x/L_c, t^* = t(L_c \alpha) \) and \( \beta = L_c \left( h/(kR_o) \right)^{0.5} \) where \( h=0.631k_o/(2R_o) \) is the heat transfer coefficient on the outer capillary surface. The solution of the dimensionless temperature is:

\[
T^*(x^*, t^*) = \frac{1}{2} \left[ 1 - \text{erfc} \left( \frac{x^*}{\beta} \right) \right].
\]

![Fig. 1. Model of the system for sample transport using thermocapillary effect.](image)
\[ T^* = \frac{1}{\beta^2} \left[ \tanh \beta \cosh(\beta x^*) - \sinh(\beta x^*) \right] + \sum_{n=1}^{\infty} D_n \exp \left\{ - \left( n - \frac{1}{2} \right)^2 \pi^2 + \beta^2 \right\} \cos \left[ \left( n - \frac{1}{2} \right) \pi x^* \right]. \]  

with
\[ D_n = 2 \int_0^{1/2} \frac{1}{\beta^2} \left[ \tanh \beta \cosh(\beta x^*) - \sinh(\beta x^*) \right] \cos \left( n - \frac{1}{2} \right) \pi x^* \, dx^*. \]

For a small temperature range, the interfacial tensions between oil and air \( \sigma_{oa} \), water and air \( \sigma_{wa} \), oil and water \( \sigma_{ow} \) are assumed to be linear functions of temperature:
\[
\sigma_{oa}(T) = \sigma_{oa,0} - \gamma_{oa}(T - T_0) \\
\sigma_{wa}(T) = \sigma_{wa,0} - \gamma_{wa}(T - T_0) \\
\sigma_{ow}(T) = \sigma_{ow,0} - \gamma_{ow}(T - T_0)
\]

where \( \sigma_{oa,0}, \sigma_{wa,0} \) and \( \sigma_{ow,0} \) are the interfacial tensions at the reference temperature \( T_0 \). The friction between the liquid plug and the capillary wall is assumed from the Hegen-Poiseuille model:
\[ \frac{dp}{dx} = \frac{8\mu}{R^2} \frac{du}{dt}. \]

where \( R \) is the radius of the plug or the inner radius of the capillary. Introducing the velocity \( u = \frac{dx}{dt} \) and the kinetic viscosity \( \nu = \frac{\mu}{\rho} \), the governing equation for the liquid/liquid plug is:
\[
\frac{du}{dt} + \left( \frac{8v_o}{R^2} + \frac{8v_w}{R^2} \frac{L_o}{L_w} \right) + \frac{2}{\rho RL_o} \left[ \sigma_{ow}(x + L_o) \cos \theta_{ow} + \sigma_{oa}(x) \cos \theta_{oa} \right] = 0
\]

In (7) the contact angles are defined relatively to the \( x \)-axis. The solution for the velocity is
\[ u = B \left[ 1 - \exp(-At) \right] \]

with
\[ A = \frac{8v_o}{R^2} + \frac{8v_w}{R^2} \frac{L_o}{L_w} \]
\[ B = - \frac{2}{\rho RL_o} \left[ \sigma_{ow}(x + L_o) \cos \theta_{ow} + \sigma_{oa}(x + L_o) \cos \theta_{oa} + \sigma_{ow}(x) \cos \theta_{oa} \right] \]

Fig. 2 illustrates qualitatively the coupling between temperature field and the movement of the liquid/liquid plug. The circles represent the positions of the plugs in space \( (x) \) and time \( (t) \).
3. Experimental results

Figure 3 describes the experimental setup. The position of the liquid/liquid plug was captured by a CCD camera. A switching device synchronizes the heater with the camera. The switching device is a miniature electromagnetic relay (Fujitsu, Takamisawa RY5W-K). The frame rate of the camera can be selected according to the plug velocity. The resolution of the camera is 640 pixels × 480 pixels. The resistive heater was made of nickel/chromium alloy (Ni80Cr20) (Goodfellow, UK). The wire has a diameter of 125 μm and is insulated by a 8-μm-thin polyimide layer. The electric current and voltage across the heaters are monitored by multimeters. Calibrated micro capillary pipettes (yellow P0799-1PAK) were purchased from Sigma-Aldrich. The capillaries are 127 mm long. The volume, outer diameter and inner diameter are listed in Table 1.

| Code  | Volume (μL) | Outer diameter (mm) | Inner diameter (mm) |
|-------|-------------|---------------------|---------------------|
| Yellow| 40          | 1.39                | 0.95                |
| Black | 20          | 1.36                | 0.60                |

Silicon oils PDMS (polydimethylsiloxane, Sigma Aldrich) was used in the experiments as the protecting liquid (- C₆H₄OSi- , ν = 100 cSt, ρ = 960 kg/m³). The experiments were carried out with a single oil plug and a oil/water plug. In the following experiments, the heating power was kept at about 2.5W. Figure 4 shows the typical movement of a oil/water plug.
Figure 5(a) compares the position versus time characteristics of a silicone oil plug in two different capillaries. The results clearly show that the same plug will move further in a smaller capillary. In an oil/water plug, the larger total plug length causes a larger friction force. Thus the oil/water plug cannot move as far as the oil plug alone.

Fig. 4. Motion of an oil/water plug

Fig. 5. Position of a silicone oil plug versus time ($\nu = 100$ cSt, $L=1.5$ mm): (a) in different capillaries, (b) with and without water plug ($\nu = 100$ cSt, $D=0.60$ mm)
4. Conclusions

We reported an actuation concept for sample transport in capillaries. In practical applications, the aqueous plugs protected by immiscible oil can be generated in a droplet-based microfluidic platform and stored in the glass capillary. Such sample plugs were successfully transported by applying heat to one end of the capillaries. The results show that the smaller the capillary, the further can the sample plug be transported. The additional water plug slows down the motion due to the larger friction force.

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