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Reciprocating thermocapillary plug motion in an externally heated capillary

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Abstract. This paper reports the concept and experimental results of reciprocating thermocapillary motion of a liquid plug in microchannels. The paper first describes an one-dimensional analytical model for the transport of micro plugs in a capillary. The model considers the coupling effect between heat transfer in the capillary wall and the surface tension driven movement of the plug. Because surface tension depends on temperature, transient temperature distribution determines the surface tension difference across a plug and thus its movement. In the experiments, we used two heaters which were activated alternatively. The liquid plug was positioned between the two heaters. The periodic temperature gradients generated by the two heaters made the liquid plug to move back and forth. The position of the plugs was captured and evaluated using a CCD camera. This paper presents the results of this motion with different switching frequencies, viscosities and plug sizes. This actuation concept has potential applications in post-processing stages for droplet-based microfluidics.

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1. Introduction

Droplet-based or digital microfluidics has been emerged as an alternative for continuous-flow microfluidics. Microdroplets formed in miniature channels can be used as a means for reagent transport and a platform for chemical reactions. Microdroplets whose lengths are larger than the channel width are considered here as microplugs. Manipulation of microdroplets or microplugs promise a huge potential in this new field of digital microfluidics (Atencia and Beebe 2005). Darhuber and Troian (2005) recently summarizes the different microfluidic actuation concepts based on the modulation of surface stresses. For instance, Pollack et al. (2000) used electrowetting effect to manipulate surface tension and subsequently to transport a droplet. Ren et al. (2002) reported a theory on the dynamic behavior of droplet transport based on electrowetting. Beside electrowetting, the surface tension gradient can also be induced by a temperature gradient. Brozoska et al. (1993) investigated the movement of a droplet on a flat surface in a temperature field. Darhuber et al. (2003) and Tseng et al. (2004) recently studied the dynamic behaviors of microdroplets in a temperature field of a flat plate. Sammarco and Burns (2000) investigated the heat transfer process inside a droplet. Yarin et al. (2004) studied the movement of droplets surrounding a thin fiber with a temperature gradient and a temperature jump as the initial condition. Recently, we reported an one-dimensional (1-D) analytical model for the transient behavior of a liquid droplet in a capillary which is heated at one end (Nguyen and Huang 2005). The same problem was investigated later by Glockner and Naterer (2005) with a two-dimensional (2-D) numerical model and microchannels fabricated in silicon.

Droplet-based microfluidics becomes increasingly popular with a number of interesting applications such as chemical analysis and protein crystallization (Song et al. 2003). Encapsulation of nanoliter droplets can also be achieved with a double emulsion, where an intermediate fluid layer works as an additional barrier between the droplet and the carrier fluid (Okushima et al. 2004; Utada et al. 2005). In the above mentioned applications, microdroplets are often generated in a microfluidic device and stored as liquid plugs in a glass capillary for storage and postprocessing (Chen et al. 2005). These liquid plugs stored inside a glass capillary can be manipulated using external heaters. The technical implementation is relatively simple and does not need microfabrication. We report in this paper a method for generating reciprocating plug motion in an externally heated capillary. The method is based on thermocapillary effect using two heaters, which are alternately activated. This method has a potential in manipulating not only the plug motion but also the flow field inside the plug. We first analytically describe the concept of this method and discuss the influence of different parameters on the motion. The peak-to-peak maximum travelling distance is taken as the evaluation base. Next, details on the experimental setup are reported. Measurement results such as plug positions and maximum travelling distance are then compared and discussed according to the different parameters such as switching frequency, liquid viscosity and plug size.

2. Actuation concept

2.1. Analytical modelling

Figure 1 depicts the model of this actuation concept. A liquid plug is positioned in a capillary between two external heaters. The capillary with two open ends is considered as long compared to the distance between the two heaters. The rest of the capillary is filled with air. The actuation concept can be described by the following coupled model. First, the transient temperature distribution in the capillary can be described with a transient 1-D heat conduction equation (Nguyen and Huang 2005). With the known temperature as a function of position and time, the temperature difference across the liquid plug and consequently the thermocapillary force can be determined. The balance equation of forces acting on the liquid plug can then be used for calculating its velocity and position as functions of time.

Since silicone oil which is used later as the plug liquid has an one order of magnitude lower thermal conductivity than glass (Table 1), we assume that the liquid plug and the glass capillary has a weak thermal interaction. Heat transfer by radiation is assumed to be negligible due to the relatively low heater temperature. Thus, the temperature distribution is assumed to be determined by the heater and the glass capillary alone:

$$\frac{\partial \theta}{\partial t} = \alpha \frac{\partial^2 \theta}{\partial x^2} - \frac{2hR_o}{\rho c(R_o^2 - R_i^2)} \theta \quad (1)$$

where θ is the temperature difference relative to the ambient temperature, R_o and R_i are the outer and inner radii of the capillary, α , ρ , and c are the thermal diffusivity, density and specific heat capacity of the capillary material, respectively. The heat transfer coefficient due to free convection at the outer surface of the capillary is estimated as $h = 0.631k_a/(2R_o)$ with k_a , the thermal conductivity of air (Yarin et al. 2002). According to Fig. 1(c), the periodic boundary conditions of the two heaters are:

$$\begin{aligned} 0 \leq t < T_h/2 : & \begin{cases} \frac{d\theta}{dx}|_{x=0} = -\frac{q_0''}{k} \\ \frac{d\theta}{dx}|_{x=D_h} = 0 \end{cases} \\ T/2 < t \leq T_h : & \begin{cases} \frac{d\theta}{dx}|_{x=0} = 0 \\ \frac{d\theta}{dx}|_{x=D_h} = \frac{q_0''}{k} \end{cases} \end{aligned} \quad (2)$$

where D_h is the distance between the two heaters, T_h is the heating period, k is the thermal conductivity of the capillary material and q_0'' is the heat flux inside the capillary wall.

For the convenience of analytical analysis, we introduce the dimensionless variables

$t^* = t/T_h$, $x^* = x/D_h$ and $\theta^* = \theta k/(q_0'' \cdot D_h)$. The dimensionless form of (1) is:

$$\eta \frac{\partial \theta^*}{\partial t^*} = \frac{\partial^2 \theta^*}{\partial x^{*2}} - \beta^2 \theta^* \quad (3)$$

with $\beta^2 = 2hD_h^2 R_o/[k(R_o^2 - R_i^2)]$, and $\eta = D_h^2/(\alpha T_h)$.

The corresponding dimensionless boundary conditions are:

$$\begin{aligned} 0 \leq t^* < 1/2 : & \begin{cases} \left. \frac{d\theta^*}{dx^*} \right|_{x^*=0} = -1 \\ \left. \frac{d\theta^*}{dx^*} \right|_{x^*=1} = 0 \end{cases} \\ 1/2 < t^* \leq 1 : & \begin{cases} \left. \frac{d\theta^*}{dx^*} \right|_{x^*=0} = 0 \\ \left. \frac{d\theta^*}{dx^*} \right|_{x^*=1} = 1 \end{cases} \end{aligned} \quad (4)$$

Solving (3) with (4) results in the dimensionless temperature:

$$\begin{aligned} \theta^*(x^*, t^*) = \Re \left\langle \sum_{n=-\infty}^{\infty} \frac{1}{\lambda_n} \left\{ \frac{F_{2n} \exp(\lambda_n) - F_{1n}}{\exp(2\lambda_n) - 1} \exp(\lambda_n x^*) + \right. \right. \\ \left. \left. + \left[\frac{F_{2n} \exp(\lambda_n) - F_{1n}}{\exp(2\lambda_n) - 1} - F_{1n} \right] \exp(-\lambda_n x^*) \right\} \exp(i2\pi n t^*) \right\rangle \end{aligned} \quad (5)$$

where $\Re$ denotes the real part of a complex number, i is the imaginary unit, and

$$\begin{aligned} n = 0 : & \begin{cases} F_{1n} = -0.5 \\ F_{2n} = 0.5 \end{cases} \\ n \neq 0 : & F_{1n} = F_{2n} = \frac{1}{2\pi n} [1 - (-1)^n] \end{aligned} \quad (6)$$

$$\lambda_n = \sqrt{\beta^2 + i2\pi n \eta}$$

The surface tension σ of the liquid depends on the temperature. For a small temperature range, a linear relation can be assumed:

$$\sigma_{lg}(\theta) = \sigma_{lg0} + \gamma(\theta - \theta_0) \quad (7)$$

where σ_{lg0} is the surface tension at the reference temperature θ_0 . The temperature field resulting from (1) is in turn a function of the position x . Assuming a weak thermal interaction between the plug and the capillary, the surface tension can be described as a function of position:

$$\sigma_{lq}(\theta) = f[\theta(x)] = g(x) = \sigma_{lg}(x) \quad (8)$$

The velocity u can be then determined through the force balance equation (Nguyen and Huang 2005):

$$\frac{du}{dt} + \left(\frac{8\nu}{R^2} \right) u + \frac{2}{\rho R_i L_{\text{plug}}} [\sigma_{lg}(x+L) \cos \phi_a - \sigma_{lg}(x) \cos \phi_r] = 0 \quad (9)$$

where L_{plug} is the length of the plug, R_i is the inner radius of the capillary, and ν is the kinetic viscosity of the plug liquid. The receding and advancing contact angles are denoted with ϕ_r and ϕ_a . The three terms in (5) represent the acceleration, the friction and the thermocapillary force, respectively.

The solution for the plug velocity is:

$$u = \frac{B}{A} [1 - \exp(-At)] \quad (10)$$

with

$$\begin{aligned} A &= \frac{8\nu}{R_i^2} \\ B &= \frac{2}{\rho R_i L_{\text{plug}}} [\sigma_{\text{lg}}(x + L_{\text{plug}}) \cos \phi_a - \sigma_{\text{lg}}(x) \cos \phi_r] \end{aligned} \quad (11)$$

At the relatively slow velocity as observed later in the experiment, we can assume the same contact angle at the receding and advancing sides ($\phi_r = \phi_a = \phi$). Applying this assumption and (7) into (10) results in

$$B = \frac{2}{\rho R_i L_{\text{plug}}} \gamma [\theta(x + L_{\text{plug}}) - \theta(x)] \cos \phi \quad (12)$$

From (10), the position of the left side of the plug can be determined as:

$$x = \int_0^t u dt = x_0 + \frac{B}{A} t + \frac{B}{A^2} \exp(-At) \quad (13)$$

The following discussion assumes a constant surface tension properties and a fixed capillary geometry (e. g. constant inner and outer radii) and focuses Only on parameters such as switching frequency, liquid viscosity and plug length.

2.2. Discussions

2.2.1. Switching frequency The properties used in this paper are listed in Table 1. Figure 2 illustrates the temperature distribution as function of the dimensionless time $t^* = t/T_h$. The “temperature surface” represents a potential surface, which causes the liquid plug to move back and forth. The results for switching frequencies $f = 1/T_h$ shows clearly that the thermal model (5) can be used for predicting the limit of the switching time. At a high frequency the temperature can not spread fast enough to cause a temperature gradient, Fig. 2(c). Consequently, the plug can not move due to the lack of thermocapillary force. The second term on the right site of (1) represents the upper critical switching frequency, which is determined by the thermal capacity of the capillary and the thermal resistance of free convection:

$$f_{\text{cr,u}} = \frac{2hR_o}{\rho c(R_o^2 - R_i^2)} \quad (14)$$

It's obvious, that for a given capillary the critical switching frequency can be increased by improving the heat transfer coefficient h , using forced convection for instance.

The lower limit of the switching frequency is determined by the plug motion. The frequency should not be too low, so that the plug is travelling out of the region of interest

between the two heaters. Applying this condition to (13) results in the equation for the lower critical switching frequency $f_{cr,l}$:

$$\frac{B}{A} \frac{1}{f_{cr,l}} + \frac{B}{A^2} \exp(-A/f_{cr,l}) - D_h = 0 \quad (15)$$

The above equation is nonlinear and can only be solved numerically.

Figure 3 depicts the typical position and velocity of a plug as functions of time. At a frequency of 0.5 Hz, no significant motion can be observed. Based on this result, the actually upper critical frequency is about one order higher than the value estimated purely from the thermal condition (14). The reason for this effect may be the fact, that the inertial energy of the plug gained by the intertidal acceleration lasts longer than the decay of the thermal field.

2.2.2. Liquid viscosity According to (9), the damping force acting on the plug is proportional to its viscosity. The higher the viscosity the smaller is the maximum peak-to-peak travelling distance. A low-viscosity plug responds quickly to the change of temperature gradient and causes an initial overshoot in velocity, Fig. 3(a). At a higher viscosity, the response is damped and the velocity curve will have a typical exponential form.

The analytical model presented here can consider the temperature dependence of the liquid viscosity. If the plug has the same temperature of the glass capillary, the viscosity values can be updated for every position. We assumed the following temperature dependency of silicone oil (Darhuber 2003):

$$\nu = \nu_0(10^{-4}T^2 - 2.62 \times 10^{-2}T + 1.57) \quad (16)$$

where ν_0 is kinematic viscosity at 25 °C. Figure 4 shows clearly the effect of temperature dependency.

2.2.3. Plug length A longer liquid plug experiences a higher temperature difference across its ends and thus a higher thermocapillary force. However, a longer liquid plug also causes more viscous friction. These two effects may compensate each other leading to independence of velocity and maximum travelling distance on the plug length. The effect of plug length on the maximum travelling distance will be investigated later in our experiments.

3. Experiments

3.1. Experimental setup and materials

Figure 5 depicts the measurement setup used for the characterization of the plug motion. The heating scheme depicted in Fig. 1(c) is based on our switching device for active micromixers reported earlier (Nguyen and Huang 2005; 2006). The switching device is a miniature electromagnetic relay (Fujitsu, Takamisawa RY5W-K). The square wave

signal from a signal generator is used for controlling the switching process. This signal can also be used for triggering the CCD camera for capturing the plug images. The frame rate of the camera can be selected according to the plug velocity. The resolution of the camera is 640 pixels $\times$ 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.

Capillary pipettes (yellow code, 40 μL) was purchased from Sigma-Aldrich. The capillaries are 127 mm long. The inner diameter and outer diameter are 0.95 mm and 1.38 mm, respectively. Silicone oils PDMS (polydimethylsiloxane, Sigma Aldrich) of different viscosities were used in the experiments as the working liquids. The oils [-Si(CH₃)₂O-, -C₇H₈OSi-, -Si(CH₃)₂O-] have different viscosities and densities but almost the same surface tension (Table 1).

3.2. Experimental results and discussions

In the following experiments, the heating power of both heaters were kept at 4.38 W by monitoring and adjusting the applied voltage. Position measurement starts when the whole system is thermally in a quasi-steady-state condition. The distance between the two heaters was $D_h = 8$ mm in all experiments. The same rate of one frame per second was kept for the CCD camera throughout all experiments. Figure 6 shows the typical images of a plug during its passage between the two heaters. The position of the left side of the plug was measured in pixels and converted into millimeters. The velocity can be evaluated using results of two subsequent frames $u = \Delta x / \Delta t$, where $\Delta t = 1$ sec is determined by the given frame rate.

Figure 7 shows the measured results of the position and velocity of a 10-mm plug at different switching frequencies. The silicone oil in use has a kinematic viscosity of 10 cSt. The results show clearly that the lower the frequency the further the plug can travel. This fact was predicted with the theoretical model described above (see Fig. 3). The over-shoot behavior of the plug velocity can be observed at a relatively low switching frequency of $f_h = 0.05$ Hz, Fig. 7(a).

Figure 8 shows the relationship between the maximum travelling distance and the switching frequency. The heat flux in the analytical model was used to fit the measured data at $f_h = 0.05$ Hz and $\nu = 10$ cSt. The results show that the maximum travelling distance decreases almost exponentially with the increasing switching frequency.

The same data in Fig. 8 is depicted in Fig. 9 to show the relationship between the maximum travelling distance and the kinematic viscosity. The results show a discrepancy between the theoretical model and the measured data. Viscous damping seems to be smaller as assumed in the analytical model. The effect of temperature dependency of viscosity may play a role here. Furthermore, the current Hagen-Poiseuille model of viscous force may overestimate the actual friction force.

Figure 10 show the relationship between the maximum travelling distance and the

plug length. The measured data was taken for a 10-cSt plug actuated at 0.05 Hz. The results show that plug length does not affect the the motion significantly. However, the trend of the measured data shows that for longer plug the viscous friction may dominate over the thermocapillary force. Thus, both the viscosity and the Hagen-Poiseuille friction model in (9) need to be revised.

4. Conclusions

We have demonstrated the periodic actuation of a liquid plug in a capillary using thermocapillary force. The reciprocating motion of the plug was investigated both analytically and experimentally. The simple relations presented in the analytical model can explain the behaviors observed in the experiments. The results show that the maximum travelling distance decreases with increasing switching frequency as well as increasing viscosity. The model can explain well the over-shoot behavior of the plug velocity as observed in the experiments. In the future work, we will focus on the influence of the capillary geometries such as inner and outer radius on the plug motion. The model of the friction force will be revised. A lubrication model may give a better explanation for the lower friction force observed in the experiments.

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Table 1. Material properties used in this work.

Materials	Glass	Silicone oil 10 cSt	Silicone oil 100 cSt	Silicone oil 1000 cSt	Air
Density $\rho(\text{kg/m}^3)$	2500	930	960	970	1.1614
Specific heat $c_p(\text{J/kgK})$	750	1150	1150	1150	1007
Thermal conductivity $k(\text{W/mK})$	1.4	0.14	0.14	0.14	2.63×10^{-2}
Thermal diffusivity $\alpha(\text{m}^2/\text{s})$	7.47×10^{-7}	1.31×10^{-7}	1.27×10^{-7}	1.26×10^{-7}	2.25×10^{-7}
Surface tension at 25°C $\sigma_0(\text{N/m})$	–	2.03×10^{-7}	2.03×10^{-7}	2.03×10^{-7}	–
Kinematic viscosity $\nu(\text{m}^2/\text{s})$	–	10^{-5}	10^{-4}	10^{-3}	2.0910^{-5}

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