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2005

Nguyen, N. T., & Huang, X. Y. (2005). Thermocapillary Effect of a Liquid Plug in Transient Temperature Fields. Japanese Journal of Applied Physics, 44(2), 1139-1142.

<https://hdl.handle.net/10356/93875>

<https://doi.org/10.1143/JJAP.44.1139>

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Thermocapillary effect of a liquid plug in transient temperature fields

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(Received)

This paper presents the theory and experimental results of thermocapillary effects of liquid plugs in an long capillary, which is exposed to a transient temperature gradient. An one-dimensional analytical model is formulated for the dynamic behavior of a liquid droplet, which is driven by the thermocapillary effect under a transient temperature field. The thermocapillary actuation concept can be used for liquid transport in microfluidics. In microfluidic applications, the temperature field is often induced by the activation of integrated heaters. The generated temperature field and temperature gradient drives a liquid droplet due to the temperature-dependent surface tension. In the initial stage, the transient temperature gradient spreads in the capillary wall much slower than the droplet itself, and thus leads to an interesting behavior of droplet motion as described in this paper. Experiments were carried out for liquid droplets with different viscosities in long glass capillaries with different radii. The capillaries are exposed to a resistive heater at one of its ends. The analytically predicted behavior of the droplet motion agrees qualitatively well with the measurement.

KEYWORDS: microfluidics, capillary, thermocapillary, surface tension, micro droplet, micro plug

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1. Introduction

Recently, microfluidics has been emerging as a key technology for biochemical analysis. Typical microfluidic applications utilize microdevices for liquid transport, mixing, separation, and chemical analysis. From many existing concepts, manipulation of micro droplets promises a huge potential in microfluidics. Pollack et al. proposed a droplet transport concept based on electrowetting, where surface tension is controlled by applied voltages.¹⁾ Another way of manipulating surface tension is utilizing the effect of thermocapillary. The temperature dependency of surface tension of a liquid/gas/solid system causes this effect. The viscosity and the surface tension of a liquid decrease with increasing temperature. A gas bubble moves against the temperature gradient toward the higher temperature, Fig. 1a. A liquid plug moves along the temperature gradient toward the lower temperature, Fig. 1b. These phenomena are also called the Marangoni-effects. In practical applications, the temperature gradient can be generated by integrated heaters.

The motion of a droplet on a flat surface was previously investigated.²⁾ Recently, this actuation concept found renewed interests from the microfluidics community.^{3,4)} These works reported dynamic behaviors of a droplet in a temperature field such as the characteristics of droplet's velocity versus its position. Besides the simulation (works done by Darhuber et al. and Tseng et al., no analytical theory was established for the dynamic behavior of these droplets.^{3,4)} In the past, a theory was reported for droplet motion caused by electrowetting.⁶⁾ Heat transfer inside a thermocapillary droplet (was studied by Sammarco et al.⁵⁾ Yarin et al.⁷⁾ reported a complex analytical model for the motion of droplets surrounding a thin fiber with temperature gradient and a temperature jump as the initial condition. However, for the best knowledge of the authors no work was done on the dynamic behavior of a liquid plug inside a capillary under a transient temperature field. In this paper we consider the one-dimensional model of a small liquid droplet in a cylindrical capillary. The model consists of two governing equations describing the spreading of thermal field in the capillary and the motion of the liquid plug.

2. Theory

2.1 Transient temperature distribution in the capillary

At a relatively low heater's temperature, the heat radiation can be neglected. The energy equation for heat transport in the capillary wall can be formulated with heat conduction and free convection as:

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

where T is the temperature difference compared to the ambient temperature, R_o is the outer diameter of the capillary, α , ρ , and c are the thermal diffusivity, the density and the specific heat capacity of the capillary's material, respectively. The initial and boundary conditions of 1 are:

$$\begin{aligned} t = 0 : \quad & T(x) = 0 \\ t > 0 : \quad & \begin{cases} x = 0, & \frac{dT}{dx} = -q'/k \\ x = L_c, & T = 0, \end{cases} \end{aligned} \quad (2)$$

where L_c , q' , k are the length of the capillary, the heat flux, and the heat conductivity of capillary's material, respectively. Introducing the dimensionless variables $T^* = T/(q' L_c/k)$, $x^* = X/L_c$ and $t^* = t/(L_c^2 \alpha)$ and:

$$\beta = \sqrt{\frac{h L_c^2}{k R_o}}$$

with $h = 0.631 k_a / (2 R_o)$ the heat transfer coefficient on the capillary's outer surface,⁷⁾ k the thermal conductivity of the capillary material, k_a the thermal conductivity of air, the dimensionless energy equation reads:

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

with the dimensionless boundary conditions:

$$\begin{aligned} t^* = 0 : \quad & T^*(x^*) = 0 \\ t^* > 0 : \quad & \begin{cases} x^* = 0, & \frac{dT^*}{dx^*} = -1 \\ x^* = 1, & T^* = 0, \end{cases} \end{aligned} \quad (4)$$

The solutions of the dimensionless temperature and temperature gradient are:

$$T^* = \frac{1}{\beta} \left[\frac{\sinh(\beta)}{\cosh(\beta)} \cosh(\beta x^*) - \sinh(\beta x^*) \right] + \sum_{n=1}^{\infty} D_n \exp \left\{ - \left[\left(n - \frac{1}{2} \right)^2 \pi^2 + \beta^2 \right] t^* \right\} \cos \left[\left(n - \frac{1}{2} \right) \pi x^* \right] \quad (5)$$

$$\frac{dT^*}{dx^*} = \left[\frac{\sinh(\beta)}{\cosh(\beta)} \sinh(\beta x^*) - \cosh(\beta x^*) \right] - \sum_{n=1}^{\infty} \left(n - \frac{1}{2} \right) D_n \exp \left\{ - \left[\left(n - \frac{1}{2} \right)^2 \pi^2 + \beta^2 \right] t^* \right\} \sin \left[\left(n - \frac{1}{2} \right) \pi x^* \right] \quad (6)$$

with

$$D_n = 2 \int_0^1 \left\{ -\frac{1}{\beta} \left[\frac{\sinh(\beta)}{\cosh(\beta)} \cosh(\beta x^*) - \sinh(\beta x^*) \right] \right\} \cos \left[\left(n - \frac{1}{2} \right) \pi x^* \right] dx^* \quad (7)$$

Figure 2 depicts the typical dimensionless temperature and temperature gradient along the capillary.

2.2 Dynamic behavior of a liquid plug in a horizontal cylindrical capillary

Figure 1b shows the model of a liquid plug in a cylindrical capillary. The surface tension is a function of the temperature, which in turn is a function of the position x if a weak thermal interaction between the plug and the capillary can be assumed:

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

The simplest model for the friction between the liquid plug and the capillary wall is the Hagen-Poiseuille model:

$$\frac{dp}{dx} = \frac{8\mu}{R^2} \frac{dx}{dt}, \quad (9)$$

where R is the radius of the plug, and μ is the dynamic viscosity of the liquid. The force equilibrium of the liquid plug can be expressed as:

$$\rho \pi R^2 L \frac{d^2 x}{dt^2} = -8\pi \mu L \frac{dx}{dt} + 2\pi R [\sigma_{lg}(x+L) \cos \theta_a - \sigma_{lg}(x) \cos \theta_r], \quad (10)$$

where L , θ_r and θ_a are the length, the contact angles at the receding and advancing ends of the liquid plug, respectively. Introducing the velocity $u = dx/dt$, rearranging (10) and introducing the kinetic viscosity $\nu = \mu/\rho$, the governing equation for the liquid plug is:

$$\frac{du}{dt} + \left(\frac{8\nu}{R^2} \right) u + \frac{2}{\rho R L} [\sigma_{lg}(x+L) \cos \theta_a - \sigma_{lg}(x) \cos \theta_r] = 0. \quad (11)$$

The three terms in the left-hand side of the above equation represent the acceleration, the friction and the surface tension, respectively. For a small temperature range, the surface tension can be assumed as a linear function of the temperature:

$$\sigma_{\text{lg}}(T) = \sigma_{\text{lg}0} - \gamma(T - T_0). \quad (12)$$

where $\sigma_{\text{lg}0}$ is the surface tension at the reference temperature T_0 . The temperature coefficient γ can be determined from the temperature function of the surface tension values. With:

$$A = \frac{8\nu}{R^2}, \quad B = \frac{2}{\rho RL} [-\sigma_{\text{lg}}(x) \cos \theta_r - \sigma_{\text{lg}}(x + L) \cos \theta_a]$$

The solution for the velocity u is:

$$u = \frac{B}{A} [1 - \exp(-At)]. \quad (13)$$

The time, position, and velocity can be non-dimensionlized according to the previous section using $t^* = t/(L_c^2/\alpha)$, $x^* = x/L_c$ and $u^* = u/(L_c/\alpha)$. The reference velocity $u_0 = L_c/\alpha$ can be considered as the diffusion speed of the temperature. Assuming the same contact angle at the receding and advancing ends θ and a short liquid plug $L \ll L_c$, the dimensionless form of (11) is:

$$\frac{du^*}{dt^{*2}} + \frac{8}{R^{*2}} \frac{\nu}{\alpha} u^* + \frac{2}{R^*} \frac{L^3 q'}{\rho k \alpha^2} \gamma \cos(\theta) \frac{dT^*}{dx^*} = 0 \quad (14)$$

With:

$$A^* = \frac{8}{R^{*2}} \frac{\nu}{\alpha}, \quad B^* = \frac{2}{R^*} \frac{L^3 q'}{\rho k \alpha^2} \gamma \cos(\theta) \frac{dT^*}{dx^*}$$

where $R^* = R/L_c$ is the dimensionless capillary radius. The temperature gradient dT^*/dx^* is taken from (6). The solutions for the dimensionless velocity u^* is:

$$u^* = \frac{B^*}{A^*} [1 - \exp(-A^* t^*)]. \quad (15)$$

Based on (15), the dynamic behavior of the liquid plug can be divided into two periods: the acceleration period and the stabilizing period. The acceleration period is determined by A^* , while the stabilizing period is determined by B^* . A less viscous plug will accelerate faster and reach a higher velocity initially. Since the liquid plug initially moves faster than the thermal diffusion, which is represented by L_c/α , the velocity then decreases due to the lower temperature gradient.

3. Experiments

3.1 Experimental setup

A measurement system was established for the observation of the dynamic behaviors of silicone oil plugs in a cylindrical capillary. The capillaries (Sigma-Aldrich) were made of glass and is 14 cm long. While the wall thickness is 200 μm , different inner radii of 1.26 mm, 1.55 mm, and 1.78 mm can be selected. One end of the capillaries is heated by a resistive wire. The resistive wire is made of Nickel/Chromium alloy (Ni80Cr20) (Goodfellow), which has a diameter of 125 μm . The wire is insulated by a 8- μm -thick polyimide layer. The heater consists of eight turns, which occupies about 3mm on the capillary. The capillary is suspended in a frame made of acrylic glass. The distance between the heater and the fixed end is $L_c = 10$ cm. The motion of the plugs was captured with a digital CCD camera. The frame rate of the camera can be selected to suit the droplet speed. The image capturing process and the activation of the heater were synchronized by an external switch. The actual experimental setup is shown in Fig. 3.

Silicone oils PDMS (poly dimethylsiloxane, Sigma-Aldrich) were used as the test liquids, which have different viscosities but almost the same surface tension. Three different oils -Si(CH₃)₂O- ($\nu = 10$ cSt, $\rho = 930$ kg/m³), -C₇H₈OSi- ($\nu = 100$ cSt, $\rho = 960$ kg/m³) and -Si(CH₃)₂O- ($\nu = 1000$ cSt, $\rho = 970$ kg/m³), were used. The position of the droplet was evaluated frame by frame using a customized program written in MATLAB. Typical images of the captured frames are shown in Fig. 4.

3.2 Experimental results and discussions

According the actual experiments, an initial plug position of $x_0^* = 0.005$ was used in the analytical results discussed in this section. The measured contact angle of 25° was also used in the analytical model. The results of the plug position versus time is depicted in Fig. 5. Figure 5a shows the theoretical position of liquid plugs at the same heat rate q' but with different viscosities of 10 cSt, 100 cSt, and 1000 cSt, respectively. The only fitting parameter is the heat rate q' . For the theoretical results, properties of the glass capillary are assumed as $\rho = 2500$ kg/m³, $c = 750$ J/m³, $k = 1.4$ W/mK, while the thermal conductivity of air is assumed as $k = 0.0261$ W/mK. The dynamic behavior of the plugs

can be seen clearly in Fig. 6, which depicts the theoretical and experimental results of velocities versus positions. The measured velocities were evaluated based on the original data of the position-versus-time measurement depicted in Fig. 5b.

The two periods of the initial behavior can be observed clearly in Fig.6. After an acceleration period, the liquid plug decelerates due to the lower temperature gradient. The acceleration period is determined by A^* in (15) or the ratio ν/α between the kinematic viscosity (momentum diffusivity) of the liquid and the thermal diffusivity of the capillary's material. The stabilizing period is determined by B^* or the heat transfer in the capillary wall as modeled in (3). A less viscous drop will accelerate faster and reach a higher velocity initially. The velocity then decreases and approaches the steady state condition.

A smaller capillary radius R^* leads to a higher values of B^* in (15), and thus a higher velocity. Although ν/α in A^* and R^* in B^* have the same order of influence on the velocity, the difference caused by ν/α can be observed clearly because of the 2 and 3 orders difference of the viscosity. In case of the capillaries used in our experiments, radii have the same size order, thus the difference caused by the capillary radius is not apparent in Fig. 7 and Fig. 8.

4. Conclusions

We have reported a simple analytical model for the transient behavior of a liquid plug in a cylindrical capillary, which is subjected to a transient temperature field. The initial condition of the temperature field exists in many practical microfluidic devices for manipulation of droplets using thermocapillary effects. A liquid plug in a capillary first accelerates to a maximum velocity under influence of the temperature gradient near a heater after switching it on. Because the thermal diffusivity is slower than the initial velocity of the plug, the plug moves out of the high-gradient region, and decelerates. The predicted behavior agrees qualitatively well with the measured results. The model can be further improved using lubrication theory for the friction term (9) and a temperature-dependent viscosity. This model can serve as a tool for designing microheaters and heating sequences for microfluidic actuation by modulation of surface tensions.³⁾

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Figure captions

Fig. 1. Movement of a gas bubble (a) and a liquid plug (b) under the thermo capillary effect

Fig. 2. Dimensionless analytical solution of (a) transient temperature distribution and (b) transient distribution of the temperature gradient along the capillary.

Fig. 3. Experimental setup for characterization of the dynamic behavior of a liquid plug in a capillary under a transient temperature field.

Fig. 4. Images of the captured frames after switching on the heater: (a) $t = 0$ sec, (b) $t = 2$ sec, (c) $t = 4$ sec.

Fig. 5. The position of liquid plugs with different viscosities after switching on the heater ($t^* \geq 0$): (a) theoretical results, (b) experimental results. The data points are measured results. The lines are fitting functions based on the presented analytical model (initial position: $x_0 = 0.5\text{mm}$, properties of the glass capillary: $\rho = 2500 \text{ kg/m}^3$, $c = 750 \text{ J/m}^3$, $k = 1.4 \text{ W/mK}$, property of air: $k = 0.0261 \text{ W/mK}$). The only fitting parameter is the heat flux $q' = 909.5 \text{ W/m}^2$, which was chosen for the droplet with a viscosity of 10 cSt. The same heat flux is applied to droplets with viscosities of 100 cSt and 1000 cSt, respectively.

Fig. 6. The velocity of liquid plugs with different viscosities at different positions: (a) theoretical results, (b) experimental results (the same conditions of Fig. 5 apply).

Fig. 7. The position of liquid plugs with different capillary radii after switching on the heater ($t^* \geq 0$): (a) theoretical results, (b) experimental results (the same conditions of Fig. 5 apply).

Fig. 8. The velocity of liquid plugs with different capillary radii and at different positions: (a) theoretical results, (b) experimental results (the same conditions of Fig. 5 apply).

FIG. 1

FIG. 2

FIG. 3

FIG. 4

FIG. 5

FIG. 6

FIG. 7

FIG. 8