A NEW COGNITION ON OSCILLATORY THERMOCAPILLARY CONVECTION FOR HIGH PRANDTL NUMBER FLUIDS

by

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A direct numerical simulations on the oscillatory thermocapillary convection in a non-axisymmetric liquid bridge of high Prandtl number fluids under normal gravity has been conducted by using a new method of mass conserving level set method for capturing any micro-scale migrations of free surface. Against the former studies, the oscillatory behaviors of surface flow (the perturbation of velocity, temperature, and free surface) and flow pattern have been quantitatively investigated simultaneously for the first time. The present results show that the instability of thermocapillary convection originates from the oscillations of velocity, temperature, and free surface at the hot corner. The velocity oscillation responds slowly to the temperature oscillation, which are opposite in transfer direction for each other, resulting in the free surface oscillation. The oscillatory thermocapillary convection in the liquid bridge is eventually ex-cited by the coupling effects of these three kinds of oscillations, which discloses clearly the oscillatory mechanism of thermocapillary convection for high Prandtl number fluids.

Key words: *oscillatory thermocapillary convection; liquid bridge; free surface*

Introduction

In air-liquid or liquid-liquid systems, the effects of evaporation, dissolution, migration of surfactant or temperature difference on the surface may lead to the emergence of surface tension gradient which induces spontaneous surface motion (such as surface deformation, the surface flow). In general, such surface motion magnified by the tractive action of surface flow is defined as Marangoni effect. In 1855, this phenomenon was first discovered and known as the *tears of wine*. Marangoni convection is one kind of natural-convection, independent of gravity. The modern space experiments and numerical studies show that when the temperature difference exceeds a certain threshold, the oscillatory thermocapillary convection appears and the symmetry steady flow transforms into the asymmetric oscillatory flow [1-3]. However, the oscillatory thermocapillary convection in melts may induce the striation formation during crystal growth and affect the quality of crystal growth. The scientific importance of this research area lies on its close relation with the flow transition and chaos in non-linear science [4, 5], which provides the theoretical basis for shallow film coating, evaporation, and floating zone crystal growth in the processing of materials [6]. Besides the experimental study in existing research means [7-10], there are mainly the linear stability analysis [11, 12], the energy stability theory [13, 14], the instability numerical simulation [15-18] and the chaotic behavior theory of

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non-linear heat convection [19]. The planned space experiment (European and Japanese Research Experiment on Marangoni Instabilities) aims at studying the Marangoni convection in liquid bridges under zero gravity conditions in order to control the hydrothermal instability. The Japan Aerospace Exploration Agency also organizes an international Marangoni convection modelling research group to investigate the oscillatory thermocapillary convection in the liquid bridge configuration determine the cause of oscillations and construct a physical model to delineate it, while the mechanism of oscillatory thermocapillary convection is still not clear so far.

The instability theory of hydrothermal wave [12, 20] considers that the oscillation of thermocapillary convection is generated by the coupling effects of thermal diffusion and thermal convection, and the oscillation has no correlation with the free surface deformation. Instead, the instability theory of the surface-wave considers that the free surface deformation plays an important role in oscillatory thermocapillary convection. Because the surface tension acts on the surface and is a restoring force, it may easily induce the disturbance along the surface [21]. The time lag between the free surface deformation and the surface temperature changes at the hot corner was investigated by the magnitude analysis [22-24]. Currently, there is no specific analysis about the contribution of various disturbances to oscillatory thermocapillary convection and its coupling mechanism. The vast majority of theoretical analyses and numerical simulations for thermocapillary convection did not consider the dynamic surface deformation or assume the free surface to be curved surface determined by the Young-Laplace equation [25, 26], which may affect the accuracy of results.

In this report, the DNS of thermocapillary convection in a liquid bridge for high Prandtl number fluid under gravity has been conducted to reveal the transition process and oscillation mechanism of thermocapillary convection by capturing the oscillatory rule of velocity, temperature, and free surface. The governing equations of thermocapillary convection under gravity are given by non-dimensional mass, Navier-Stokes and energy conservation equations and solved on a staggered grid. The key parameters are Prandtl number, Reynolds number, Capillary number, and Marangoni number. The advection terms are discretized by the quadratic upstream interpolation for convective kinematics method and the other terms by the fourth order central finite-difference method. The second-order Adams-Bashforth method is used as the time integration scheme. The continuum surface force model is employed to treat the surface tension force on the surface. The present model of thermocapillary convection in the liquid bridge has been confirmed not only by our previous articles [27-29] but also by studies of other scholars [30, 31]. Moreover, the surface movement induced by the thermocapillary convection in the liquid bridge of high Prandtl number fluid can be extremely small, in the order of microns. It is not easy to capture such tiny surface movements in gas-liquid two-phase flow simulations, especially when the surface displacement is smaller than one mesh spacing. The Volume of Fluid method would have an insurmountable difficulty in overcoming this spatial resolution problem. In contrast, the level set approach can avoid this problem using the following algorithm developed in the present work.

Physical and mathematical models

The liquid bridge with radius, R, and height, H, is suspended between two coaxia disks and surrounded by the air in a rectangular container with height, H, an d width, $4R$, as shown in fig. 1(a). The radius and height of liquid bridge are R = 0.5 mm and, H = 0.5 mm, respectively. The temperature difference between the two disks is $\Delta T = T_t - T_b$, where T_t and T_b are the temperature of the upper and bottom disks, respectively. The general governing equations of the problem under gravity are given by the following non-dimensional mass, Navier-Stokes and energy conservation equations:

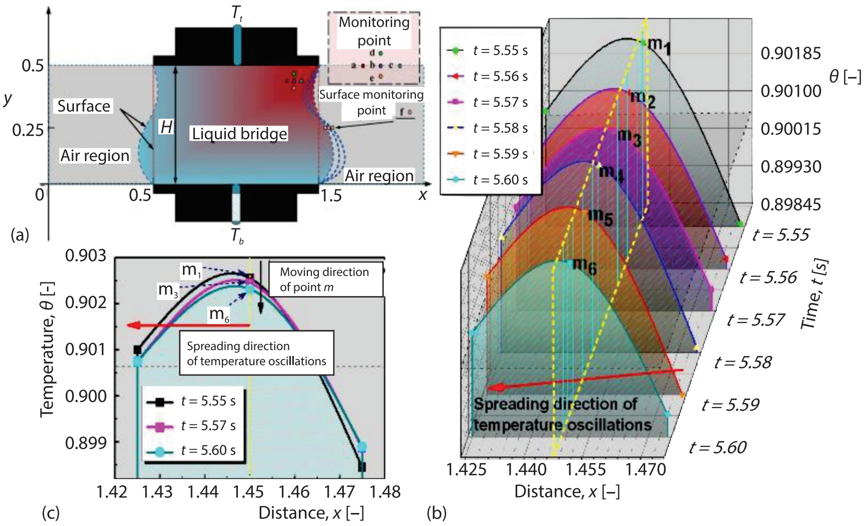

Figure 1. (a) Schematics of non-axisymmetric liquid bridge model, *a*, *b*, *c*, *d*, *e* are five monitoring points at hot corner, and *f* is a surface monitoring point at mid-height of free surface, (b) the 2-D view of temperature propagation within point *a* to *c* in transversal direction at 5.55, 5.57, and 5.6 seconds, and (c) the 3-D view of temperature propagation within point *a* to *c* in transversal direction from 5.55-5.6 seconds

$$\mathbf{u}_t + (\mathbf{u}\nabla)\mathbf{u} = \mathbf{g} + \frac{1}{\rho}\left\{-\nabla p + \frac{1}{\mathrm{Re}}\nabla(2\mu\mathbf{D}) + \left[\frac{1}{\mathrm{We}} - \frac{\mathrm{Ca}}{\mathrm{We}}\theta\right]\zeta\delta(d)\mathbf{n}\right\} \tag{1}$$

$$\nabla\mathbf{u} = 0 \tag{2}$$

$$\frac{\partial\theta}{\partial t} + \nabla(\mathbf{u}\theta) = \frac{1}{\mathrm{Ma}}\nabla^2\theta \tag{3}$$

where $\mathbf{u} = (u, v)$ is the fluid velocity, $\rho = \rho(\mathbf{x}, t)$ – the fluid density, $\mu = \mu(\mathbf{x}, t)$ – the fluid viscosity, $\mathbf{D}$ – the viscous stress tensor, ξ – the curvature of the interface, d – the normal distance to the interface, δ – the Dirac delta function, $\mathbf{n}$ – the unit normal vector at the interface, and t – the dimensionless time. The surface tension coefficient is considered to be a linearly function of temperature and defined as $\sigma = \sigma_c + \sigma_T(T - T_b)$, where σ_c is a reference value of surface tension, σ_T – the temperature coefficient of surface tension. We denote $\sigma'_T = \partial\sigma/\partial T$, T – the temperature, and $\mathbf{g}$ – a body force induced by a lateral acceleration.

The level set method was originally introduced by Osher and Sethian [32] to numerically predict the moving interface $\Gamma(t)$ between two fluids. Instead of explicitly tracking the interface, the level set method implicitly captures the interface by introducing a smooth signed distance from the interface in the entire computational domain. The level set function $\phi(\mathbf{x}, t)$ is taken to be positive outside the liquid bridge, zero on the interface and negative inside the liquid bridge. The interface motion is predicted by solving the convection equation for the level set function of $\phi(\mathbf{x}, t)$:

$$\phi_t + \mathbf{u}\nabla\phi = 0 \tag{4}$$

The density ρ and viscosity μ in the flow field can then be expressed:

$$\rho(\phi) = 1 + \left(\frac{\rho_g}{\rho_l - 1}\right) H_\alpha(\phi) \tag{5}$$

$$\mu(\phi) = 1 + \left(\frac{\mu_g}{\mu_l - 1}\right) H_\alpha(\phi) \tag{6}$$

$$H_\alpha(\phi) = \begin{cases} 0 & \phi < -\alpha \\ (\phi + \alpha)/(2\alpha) + \sin(\pi\phi/\alpha)/(2\pi) & |\phi| \le \alpha \\ 1 & \phi > \alpha \end{cases} \tag{7}$$

where α is the prescribed thickness of the liquid-gas interface, and we used $\alpha = 2.0\Delta x$ in the present work where Δx is the grid step spacing in x-direction.

The surface movement induced by a small vibration and the thermocapillary convection in the liquid bridge of high Prandtl number fluid can be extremely small, while it can have a non-negligible effect on the thermocapillary convection. Moreover, due to the reduction in the thermal boundary-layer thickness and its implications for finer grid resolution in the numerical analyses for high Prandtl number fluids, it is not easy to capture such tiny surface movements, especially when the surface displacement is smaller than one mesh spacing. Therefore, an algorithm catching the surface displacement smaller than one mesh spacing was developed in the present work based on the level set method and described below:
– *Step 1*. Determine a monitoring point on the interface.
– *Step 2*. Find out the grid point, which bounds the monitoring point outside the interface or on the interface when the interface moves through the grid at every time step.
– *Step 3*. Calculate the position of the monitoring point using the information of the grid point found out in *Step 2*.

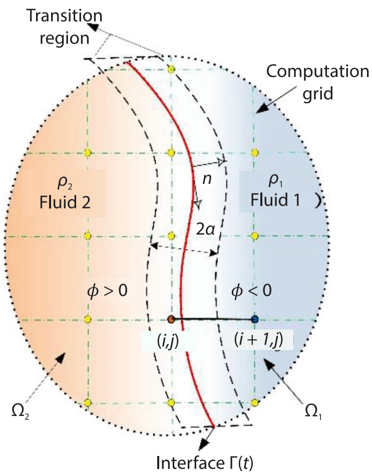

Figure 2. An interface intersecting with a grid line

Let us take a monitoring point on the interface of the liquid bridge. There are two ways for the interface to intersect with the grid. One way is that the interface intersects with the grid line between two grid points as shown in fig. 2, and the other is that the interface directly falls on one of the grid points. In fig. 2, (i, j) and $(i + 1, j)$ represent two arbitrary grid points on opposite sides of the interface. The level set function ϕ can be constructed by choosing the sign of ϕ to be negative inside the interface and positive outside the interface. If the interface intersects with the grid point, for example, (i, j), the level set function ϕ at this point should be zero and the position of the monitoring point can be determined by using the co-ordinate value of (i, j). If the interface intersects with the grid line between the two grid points, for example, (i, j) and $(i + 1, j)$, the two points should be determined:

$$\phi(i, j)\phi(i+1, j) < 0 \tag{8}$$

According to the principle of the level set method, the value of the level set function ϕ at (i, j) is equal to the signed distance from this point (i, j) to the interface. Therefore, the position of the monitoring point can be determined by using the co-ordinate value of (i, j) and the value of the level set function ϕ at (i, j). The surface displacement smaller than one mesh spacing can be captured according to the present algorithm, which allows us not to have to use the finer grid resolution in the numerical analyses for high Prandtl number fluids.

The flow in the liquid bridge and the ambient air were both assumed to be steady initially. All walls of the computational region were assumed to be adiabatic except for the hot and cold disks of the liquid bridge, which were maintained at constant temperature T_t and T_b, respectively, with a temperature difference ΔT. Consequently, the boundaries conditions would be satisfied:

$$\Theta = 0, \ (y = 0) \tag{9}$$

$$\Theta = 1.0, \ (y = 1.0) \tag{10}$$

In the two-phase system studied here, the initially stationary liquid bridge was considered with the initial velocity for both liquid bridge and ambient air:

$$\mathbf{u}(t = 0) = 0 \tag{11}$$

The non-slip condition was used for all walls of the computational domain:

$$\mathbf{u} = 0 \tag{12}$$

In order to verify the present model, a comparison between the present results and available numerical results is performed by prescribing a liquid bridge of 10 cSt silicone oil. The results match those of Shevtsova *et al.* [33] well, as shown in fig. 3. Moreover, the preciseness of our present calculation model has been fully verified by other scholars, see fig. 3. in [34, 35].

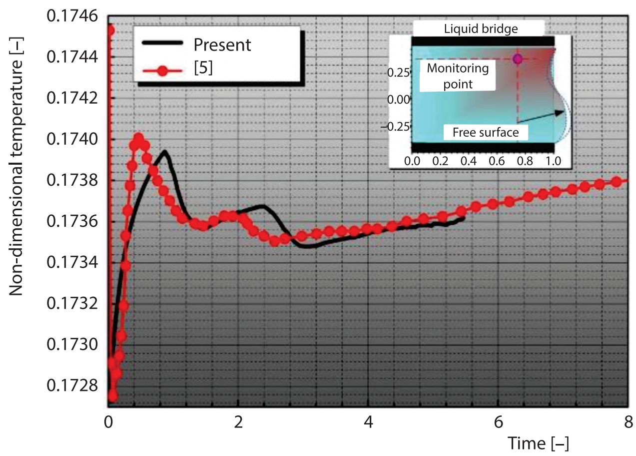

Figure 3. Time dependences of the temperature at the point (x = 0.75, y = 0.325, Pr = 10^5, Re = 229.4, g = 9.81 m/s^2, ΔT = 40 K, H/R = 4/3, D = 6 mm)

A liquid bridge of 10 cSt silicone oil is considered under normal gravity in the present paper. The temperature difference between the top and bottom disks is ΔT_0 = 25 K, and other initial computational conditions adopted are: Pr =111.67, Ma = 1.3, Ca = 0.075. The values of the parameters are listed in tab. 1. Five monitoring points are set at the hot corner of liquid bridge, and their dimensionless locations are a(1.425, 0.475), b(1.45, 0.475), c(1.475, 0.475), d(1.45, 0.4875), e(1.45, 0.425), respectively, as shown in fig. 1(a).

Table 1. Physical properties of 10 cSt silicone oil

		Silicone oil
Density ρ, (25 °C)	[kgm^{-3}]	935
Dynamic viscosity μ, (25 °C)	[Nsm^{-2}]	$9.35 \cdot 10^{-3}$
Kinematic viscosity ν, (25 °C)	[m^{2}s^{-1}]	10^{-5}
Thermal diffusivity α, (25 °C)	[m^{2}s^{-1}]	$8.96 \cdot 10^{-8}$
Thermal conductivity κ , (25 °C)	[Wm^{-1}K^{-1}]	0.14
Surface tension σ, (25 °C)	[Nm^{-1}]	$20.1 \cdot 10^{-3}$
Temperature coefficient of surface tension σ_T	[Nm^{-1}K^{-1}]	$-6.83 \cdot 10^{-5}$
Temperature coefficiemt of viscosity ν_T	[–]	0.55
Specific heat capacity, C_p	[Jkg^{-1}K^{-1}]	1672

Results and discussion

Figure 4 shows the temperature oscillations at five monitoring points at the hot corner. The temperature oscillations are periodic ones at five monitoring points, and the average oscillating period is about t = 0.04 seconds. The earliest onset of oscillation is roughly at t = 5.52 seconds at point c, and the corresponding critical temperature difference is ΔT_{cr} = 57.82 K. It is found that the temperature oscillation first appears at the point c compared with those at other monitoring points, and the oscillation onset at point b is about 0.01 seconds earlier than that at point a. Because of violent coupling effects of the ambient temperature and the heat conduction near the free surface, the temperature at point c first loses stability, showing that the temperature oscillation originates from the position close to the free surface. From the investigation on the amplitude, the temperature oscillation at point c is the strongest and the oscillating peak increases continuously. The valley of temperature oscillation declines with time at point a and the amplitude tends to expand. The oscillatory patterns of the temperature at point b, d, and e are basically the same, and the peak remains constant and the amplitude tends to expand.

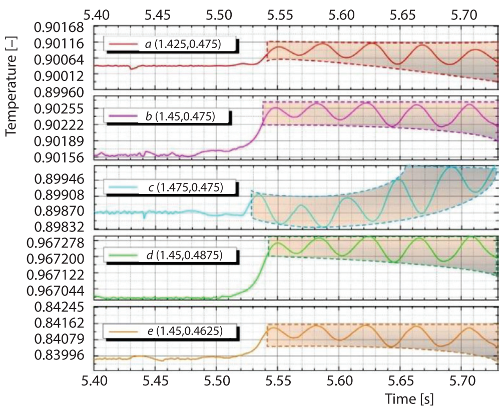

Figure 4. Temperature oscillations at *a*, *b*, *c*, *d*, and *e* five monitoring points shown in fig. 1(a), respectively

Figure 4 shows that the temperature oscillation exists at both transversal and vertical monitoring points, that is, it transfers along both transversal and vertical directions. In the vertical direction, the location of monitoring points d, b, and e are successively away from the hot disk, and the disturbance energy transfers from upward to downward. In the transversal direction, the early stage (t = 5.55-5.60 seconds) of oscillation is investigated to reveal the propagation pattern of temperature oscillation, fig. 1(b). The wave moves down from position m_1 to m_6 (see the direction of black arrow in fig. 1(c), therefore, the successive attenuation of the amplitudes shows that the direction of energy consumption is from the free surface to the inner of liquid bridge, see the direction of red arrow in fig. 1(b). The point m_1 moves down from at the position b in the waves, see the direction of black arrow in fig. 1(b), therefore, the successive attenuation of the amplitudes shows that the direction of energy consumption is from the free surface to the inner of liquid bridge, see the direction of red arrow in fig. 1(b). In addition, the oscillatory types of temperature are the same in transversal and vertical directions, $c \rightarrow b \rightarrow a$, $d \rightarrow b \rightarrow e$, see fig. 3.

Figures 5 and 6 show the oscillations of transverse and vertical velocities at five monitoring points shown in fig. 1(a). The average oscillating period of transversal and vertical velocities at five monitoring points is about t = 0.04 seconds. The earliest onset time for the velocity oscillation is about t = 5.53 seconds, which has a time lag (t = 0.01 seconds) compared with that of temperature oscillation (t = 5.52 seconds). From figs. 4-6, two peculiar physical phenomena are found:

- The temperature oscillation precedes the velocity oscillation.
- The oscillatory patterns of transversal and vertical velocities are similar, while the oscillatory intensity of transversal velocity is larger than that of vertical oscillation.

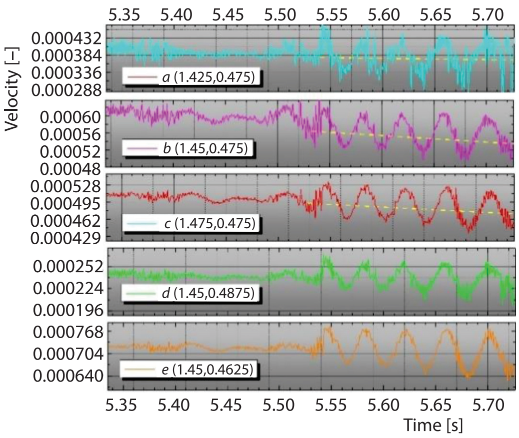

Figure 5. Oscillation of transverse velocity, *u*, at five monitoring points at hot corner

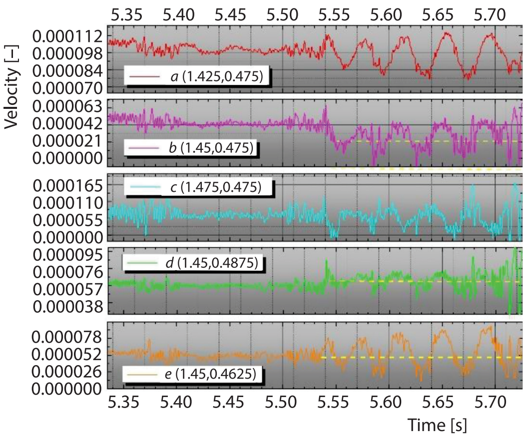

Figure 6. Oscillation of vertical velocity, *v*, at five monitoring points at hot corner

Based on the attenuation direction of the velocity amplitude in figs. 4 and 5, it is found that the propagation direction of transversal velocity oscillation is from inner to surface, $\bar{A}_a = 4 \cdot 10^{-5} \rightarrow \bar{A}_b = 3.45 \cdot 10^{-5} \rightarrow \bar{A}_c = 2.75 \cdot 10^{-5}$ in fig. 5, which is from bottom to top for the vertical velocity oscillation, $\bar{A}_e = 2.93 \cdot 10^{-5} \rightarrow \bar{A}_b = 0.975 \cdot 10^{-5} \rightarrow \bar{A}_d = 0.775 \cdot 10^{-5}$ in fig. 6. Therefore, it is found, for the first time, that the propagation directions of velocity and temperature oscillations are opposite. The velocity oscillations are mainly affected by variations of thermocapillary convection and the movement of the cell flow. However, the temperature oscillation is directly affected by the thermal disturbance at the hot corner. Eventually, the response of the velocity oscillation the temperature oscillation is slow. Therefore, it can be concluded that the mechanism of velocity and temperature oscillations is different. The difference and complex coupling effects of these two oscillations cause the fluctuation of free surface at the hot corner and the instability of thermocapillary convection.

Figure 7 shows the flow structure of thermocapillary convection within an oscillatory period from t_1 = 5.54-5.58 seconds. First, the expansion of right cell flow extends to the left side, and the outermost streamline of right cell flow has crossed the central location (y = 1.00) at t_1, indicating that the peripheral fluid of right cell flow has invaded the left cell flow, fig. 7(a). The influence of right cell flow is weakened as the development of thermocapillary convection from t_1 to t_2, and the interaction of cell flows on both sides reaches equilibrium at t_2, fig. 7(b). From t_2 to t_3, the left cell flow expands into the right cell flow, and the outermost streamline of left cell flow has crossed the central location, ($y = 1.00$) at t_3, fig. 7(c).

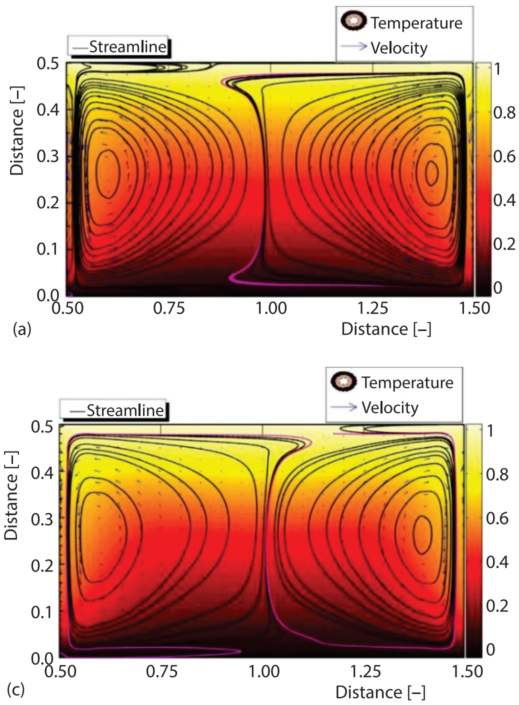

Figure 7. Flow stream in the liquid bridge at different times; (a) t_1 = 5.54 seconds, (b) t_2 = 5.56 seconds, and (c) t_3 = 5.58 seconds; the alternative expansion and encroachment of left and right cell flows are shown by using the labeled purple outermost streamline of cell flow

Figure 8 illustrates the oscillations of free surface, temperature, transversal and vertical velocities at the surface monitoring point *f*. The temperature first oscillates regularly, and then the velocity and the free surface start to oscillate. The surface fluctuation is sensitive to the temperature one, and the asynchronous characteristic between temperature and velocity oscillations excites the surface oscillation. The oscillating periods of surface and transversal velocity are basically the same, which are high frequency oscillations compared with those of temperature and vertical velocity. The onset times of temperature and velocity oscillations at the hot corner are about 0.01 second ahead of those at mid-height surface, respectively, as shown in figs. 4-6, and figs. 8(b) and 8(d), which further confirms that the oscillatory thermocapillary convection is generated first at the hot corner. The oscillating frequency of transversal velocity at point *f* is higher than those at five monitoring points *a-e* as shown in figs. 8(c) and 5, showing that the oscillation of transversal velocity at mid-height point is different with those at the hot corner. At the same time, the oscillatory frequency of temperature is smaller compared with those of transversal velocity and free surface at the monitoring point *f* as shown in fig. 8.

A new oscillatory mechanism model of thermocapillary convection for the high Prandtl number fluid is elaborated based on the aforementioned analyses, fig. 9, in the present report. The region near the free surface at the hot corner (active region) is the most sensitive region affected by the variation of temperature and flow, and the disturbance in this region affects the entire flow in the liquid bridge. Although the surface size of active region is smaller compared with that of whole surface, the velocity of surface flow in the active region is large. The

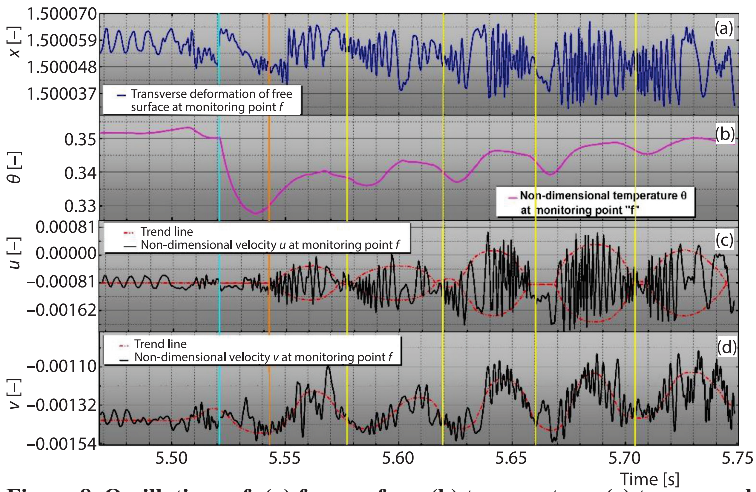

Figure 8. Oscillations of; (a) free surface, (b) temperature, (c) transversal velocity, and (d) vertical velocity at the mid-height surface point *f* shown in fig. 1(a)

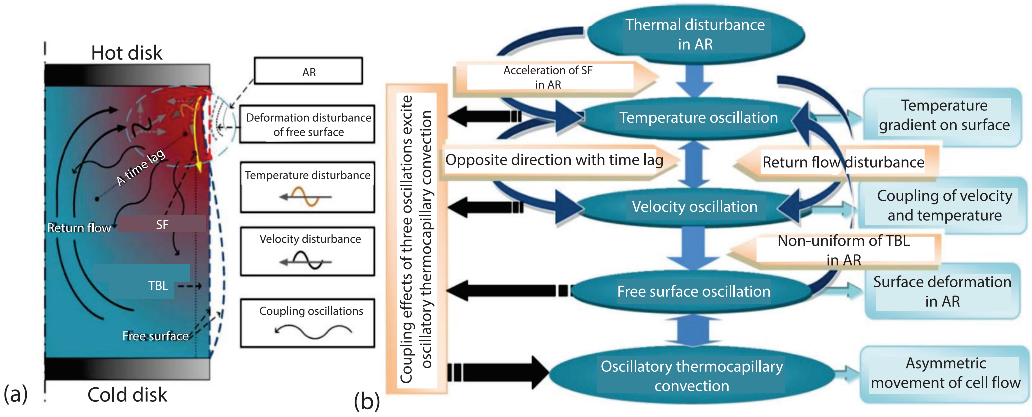

Figure 9. (a) and (b) Oscillatory mechanism model of thermocapillary convection; the region near the free surface at the hot corner is referred to as the active region, which is abbreviated to active region (AR); the surface flow is abbreviated to surface flow (SF), and the thermal boundary-layer is abbreviated to thermal boundary-layer (TBL)

time scale in this region is smaller than that of entire convection in the liquid bridge. Further, the disturbance in the active region occurs first. Obviously, the smaller time scale in the active region means that the violent variation of temperature and surface flow occurs, which is relative to the internal return flow, and a time lag between the surface flow in the active region and the returning flow occurs. Specifically, the pressure gradient variation in the active region due to accelerated surface flow results in the surface deformation. Moreover, the thermal boundary-layer is thin for the high Prandtl number fluid, which leads to the increasing of temperature gradient along the free surface. At the same time, the velocity of surface flow is improved, which accelerates the cooling of active region, and the temperature gradient induces the periodic oscillation of temperature in the active region. On the other hand, the supplementary surface flow is driven to the active region by the internal return flow, which cools the active region and brings disturbance information of inner convection and movement of vortex center. Therefore, there is a response lag between the oscillations of velocity and temperature. Together with the opposite propagation direction for velocity and temperature oscillations, the oscillation of free surface is ex-cited. The coupling effects of three oscillations eventually induce the asymmetric oscillation of thermocapillary convection. At the same time, the analysis for the direction of energy transmission further confirms that the oscillation of thermocapillary convection originates from the hot corner.

Conclusions

In summary, the conclusions are as follow.

- The velocity oscillation is mainly affected by the interior flow variation in the liquid bridge, and there is a time lag in space between the surface flow and the returning flow. The temperature oscillation is directly affected by the thermal disturbance at the hot corner, and the velocity oscillation responds slowly to the temperature oscillation.
- The propagation directions of velocity and temperature oscillations are opposite, and the difference and complex coupling effects of two oscillations intensify the oscillation of free surface.
- The instability of thermocapillary convection results from the interaction of free surface, velocity and temperature oscillations, and the coupling effects of three oscillations induce the asymmetric flow of thermocapillary convection.
- Comparing the onset time for the oscillations at the hot corner with that at the midheight of free surface, it is found that the oscillation of thermocapillary convection originates from the hot corner and the high frequency oscillation exists at the midheight of surface.
- In this paper, a new mechanism model for the oscillatory thermocapillary convection for high Prandtl fluids is summarized and some peculiar phenomena are found for the first time. These findings are useful not only to the research on the thermocapillary convection but also to the theoretical basis for the non-linear dynamic system control and the study on complex behavior in chemical and biological systems.

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Nomenclature

$\bar{A}$ – amplitude, [–]
Ca – Capillary number (= $|\sigma_T'|\Delta T/\sigma$), [–]
g – acceleration of gravity, [ms^{-2}]
H – height of liquid bridge, [m]
L – characteristic length (=2R), [m]
Ma – Marangoni number (= $|\sigma_T'|\Delta TL/\rho\nu\kappa$), [–]
Pr – Prandtl number (= ν/κ),[–]
p – dimensional pressure, [Nm^{-2}]
R – radius of liquid bridge, [m]
Re – Reynolds number (=$|\sigma'|\Delta TL/\rho\nu^2$), [–]
T – Temperature, [K]
t – time, [s]
U – characteristic velocity (=$|\sigma_T'|\Delta T/\mu$), [–]
u – dimensionless transverse velocity (=u_s/U), [–]
u_s, v_s – vertical velocity, transverse velocity, [ms^{-1}]
v – dimensionless vertical velocity (=v_s/U), [–]
We – Weber number (= U^2L/σ),[–]
X, Y – dimensionless co-ordinates ($X = x/L$, $Y = y/L$), [–]
x, y – dimensional co-ordinates,[m]

Greek symbols

α – thermal diffusivity, [m^{2}s^{-1}]
θ – dimensionless temperature [$= (T - T_b)/(T_t - T_b)$], [–]
κ – thermal conductivity, [Wm^{-1}K^{-1}]
ν – kinematic viscosity, [m^{2}s^{-1}]
μ – dynamic viscosity, [Nsm^{-2}]
ρ, ρ_0 – density, reference density, [kgm^{-3}]

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