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Oscillatory instability of Rayleigh–Marangoni–Bénard convection in two-layer liquid systems

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Abstract

The oscillatory behaviour of the Rayleigh–Marangoni–Bénard convective instability (R-M-B instability) regarding two combinations of two-layer fluid systems has been investigated theoretically and numerically. For the two-layer system of Silicone oil (10cSt) over Fluorinert (FC70), both linear instability analysis and 2D numerical simulation show that the instability of the system depends strongly on the depth ratio $H_r = H_1/H_2$ of the two-layer liquid. The oscillatory regime at the onset of R-M-B convection enlarges with reducing $\Gamma = Ra/Ma$ values. In the two-layer system of Silicone oil (2cSt) over water, it loses its stability and onsets to steady convection at first, then the steady convection bifurcates to oscillatory convection with increasing Rayleigh number Ra. This behaviour was found through numerical simulation above the onset of steady convection in the case of $\Gamma = 2.9$, $\varepsilon = (Ra - Ra_c)/Ra_c = 1.0$, and $H_r = 0.5$. Our findings are different from the previous study of the Rayleigh–Bénard instability and show the strong effects of the thermocapillary force at the interface on the time-dependent oscillations at or after the onset of convection. We propose a secondary oscillatory instability mechanism to explain the experimental observation of Degen et al. [Phys. Rev. E, 57 (1998), 6647–6659].

1. Introduction

The convective instabilities and mechanisms in two or more superposed layers of liquid–liquid systems are more complex than those in the single-layer

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systems, due to the competition between instabilities in the separate layers and the various interfacial surface tension driven modes. Many scientists have extensively studied two- or multiple-layer convection in view of several interfacial phenomena in nature and in numerous engineering applications. The study of two-layer convection becomes an important new direction for the field of pattern formation and bifurcation phenomena in non-equilibrium systems, and much attention has been focussed on the instability analysis of multi-layered convection in the case of external thermal gradient perpendicular to the liquid interface, for example the classic problems of the Rayleigh–Bénard convection [1–4] or the Rayleigh–Marangoni–Bénard convection [5–9] in two-layer liquids.

For the flow in a two- or multi-layer system, one of the most interesting problems is the possibility of finding time-dependent states at or after the onset of convection, since the buoyancy-induced oscillatory instability was discovered by Gershuni and Zhukhovitskii [1]. The oscillatory convection in the two-layer Rayleigh–Bénard system where thermocapillarity is negligible was investigated theoretically [1, 2, 4] and experimentally [2, 10]. Both, the instability analysis and experimental observation found two possible convective states: thermal coupling or mechanical coupling in two-layer Rayleigh–Bénard convection for different combinations of two liquids. In a narrow transition region between the two different states, the time-dependent convection (the Hopf modes) may appear [11]. Renardy and Joseph [3] have conducted fairly extensive analytical studies on the stability of the two-layer Bénard system by using the perturbation theory. Their findings indicate that the onset of instability could be oscillatory. The linearised perturbation analysis of the system performed by Rasenat et al. [2] reveals that oscillatory instability is possible due to the cyclic variation between viscous and thermal coupling. Colinet and Legros [4] revisited the problem theoretically by assuming an undeformable interface and by selecting the fluid properties of a model two-layer system, and gave a typical stability diagram for one range of layer depth ratios in which the oscillatory modes arise between the two different stationary convective states.

Recently, experiments on the two-layer Rayleigh–Bénard system with two different pairs of fluids were performed by Degen et al. [10]. They found time-dependent patterns at or near the convective onset, but some evident differences such as the periods of the time-dependent flow and the time-dependent region of layer depth ratios have also been shown in comparison with the theoretical predictions [11]. In fact, for the two-liquid systems used in Degen's experiments, the oscillatory convection region for the total layer depth $H = 12$ mm is too small to observe experimentally and to confirm the oscillatory instability phenomena at the convective onset. It should be mentioned that most previous investigations of this problem were performed

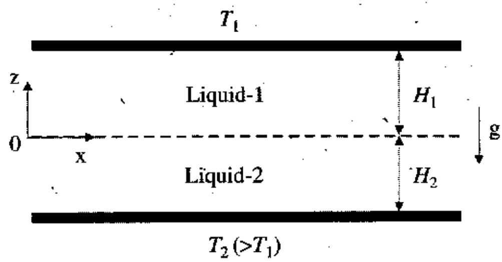


Figure 1 Schematic diagram of two-layer liquids.

mainly with regard to the instability behaviour induced by the buoyancy force, focussing on the oscillatory behavior at onset of the whole system. Nepomnyashchy and Simanovskii pointed out that the Marangoni effect could change the supercritical oscillatory instability in the pure buoyancy-driven case into a critical one [9].

The objective of the present study is to investigate theoretically the interaction between Rayleigh–Bénard instability and Marangoni instability in a two-layer system with the emphasis of the oscillatory instability at or after the onset of the Rayleigh–Marangoni–Bénard convection.

2. Physical model and basic equations

The theoretical model of a two-layer Rayleigh–Marangoni–Bénard system is assumed to be infinite in the horizontal direction as shown schematically in Figure 1. The total depth of two layers, $H = H_1 + H_2$, is used as the non-dimensional scale for length, where the subscripts 1 and 2 refer to the upper and the lower fluid layers, respectively. The thickness ratio of two layers is defined as $H_r = H_1/H_2$. A temperature difference $\Delta T = T_2 - T_1$ is imposed parallel to the acceleration of gravity **g** between the top and bottom isothermal rigid plates. When $\Delta T > 0$, the bottom boundary is hotter than the top boundary $(T2 > T1)$. The dimensionless ratios of the fluid properties are $\kappa^* = \kappa_1/\kappa_2$ (thermal diffusivity), $\beta^* = \beta_1/\beta_2$ (coefficient of thermal expansion), $\chi^* = \chi_1/\chi_2$ (thermal conductivity), $\mu^* = \mu_1/\mu_2$ (dynamic viscosity), $\rho^* = \rho_1/\rho_2$ (density), and $\nu^* = \nu_1/\nu_2$ (kinematical viscosity), respectively. The interface between the immiscible liquids is assumed to be flat [9, 16]. The interfacial tension at the interface is considered to be a linear function of temperature: $\sigma = \sigma_0 + (\partial\sigma/\partial T)(T - T_0)$, where T_0 is the reference temperature of the interface and $\partial\sigma/\partial T$ is usually negative.

The governing equations for each fluid layer are the heat transport equation and the Navier–Stokes equations with the Boussinesq approximation, i.e., only the densities ρ_i are dependent on the temperature, $\rho_i = \rho_{0i}[1 - \beta_i(T_i - T_0)]$. In a two-layer Rayleigh–Marangoni–Bénard system, the convection arises due to buoyancy and temperature dependence of the interfacial tension, and their contributions are estimated by two important non-dimensional parameters: the Rayleigh number

$$Ra = g\beta_2 \Delta T H^3/(\nu_2 \kappa_2),$$

and the Marangoni number

$$Ma = (-\partial\sigma/\partial T)\Delta T H/(\mu_2 \kappa_2).$$

At the onset of convection, these parameters correspond to the critical values Ra_c, Ma_c with the critical temperature difference ΔT_c. By using ν_2/H, H^2/ν_2, H, and ΔT as the scaling factors for velocity, time, length, and temperature, respectively, the dimensionless governing equations in such two-layer systems are

$$\nabla \cdot \mathbf{V}_i = 0, \tag{1}$$

$$\frac{\partial \mathbf{V}_i}{\partial t} + \mathbf{V}_i \cdot \nabla \mathbf{V}_i = -C_i^{\rho} \nabla p_i + C_i^{\nu} \nabla^2 \mathbf{V}_i + C_i^{\beta} \mathbf{g}\theta, \tag{2}$$

$$\frac{\partial \theta_i}{\partial t} + \mathbf{V}_i \cdot \nabla \theta_i = -C_i^{\kappa} \nabla^2 \theta_i, \tag{3}$$

where $i = 1$ identifies the upper-layer liquid; $i = 2$, the lower-layer liquid. $\mathbf{V}_i = (u_i, 0, w_i)$ is the dimensionless velocity; $\mathbf{g}$ is gravitational acceleration; $\theta_i = (T_i - T_0)/\Delta T$, dimensionless temperature; and p_i, dimensionless pressure. The constants in the right-hand side of the dimensionless equations (1)–(3) above are respectively: $C_1^{\rho} = 1/\rho^*$, $C_1^{\nu} = \nu^*$, $C_1^{\beta} = Ra\beta^*/Pr$, $C_1^{\kappa} = \kappa^*/Pr$; $C_2^{\rho} = 1$, $C_2^{\nu} = 1$, $C_2^{\beta} = Ra/Pr$, $C_2^{\kappa} = 1/Pr$, where $Pr = \nu_2/\kappa_2$ is the Prandtl number corresponding to the physical properties of liquid (2). Boundary conditions and the initial condition needed in numerical simulation can be found in [13].

For the linear instability analysis of the problem in such a two-layer system, we considered the base state of the system with a flat interface at $z = 0$, a zero velocity field, and a temperature field that varies linearly with z in each fluid. Introducing the spatial normal perturbations proportional to $exp[\lambda t + i(k_x x + k_y y)]$ into the linearised form of the governing equations (1)–(3), the dimensionless linearised governing equations of the two-layer system can be written for the amplitudes of the perturbation quantities w_i, the velocity component in the vertical direction z and θ_i, the temperature in each layer [14]:

$$\nu^*(D^2-k^2)^2 w_1 - \frac{Ra}{Pr}\beta^* k^2\theta_1 = \lambda(D^2-k^2)w_1, \tag{4}$$

$$k^*(D^2-k^2)\theta_1 - \frac{\partial T_1}{\partial z} Pr w_1 = \lambda Pr \theta_1, \tag{5}$$

$$(D^2-k^2)^2 w_2 - \frac{Ra}{Pr}k^2\theta_2 = \lambda(D^2-k^2)w_2, \tag{6}$$

$$(D^2-k^2)\theta_2 - \frac{\partial T_2}{\partial z} Pr w_2 = \lambda Pr \theta_2, \tag{7}$$

with the boundary conditions

$$w_1 = Dw_1 = \theta_1 = 0 \quad at\ z = \frac{H_r}{1+H_r}, \tag{8}$$

$$w_1 = w_2 = 0, \quad Dw_1 = Dw_2,$$

$$\theta_1 = \theta_2, \quad \chi^* D\theta_1 = D\theta_2,$$

$$D^2 w_2 - \mu^* D^2 w_1 = -\frac{Ma}{Pr}k^2\theta_2 \quad at\ z = 0, \tag{9}$$

$$w_2 = Dw_2 = \theta_2 = 0 \quad at\ z = -\frac{1}{1+H_r}, \tag{10}$$

where D is the dimensionless differential operator d/dz, λ is the time growth rate, $k = (k_x^2 + k_y^2)^{1/2}$ is the dimensionless wave number, and $\partial T_i/\partial z$ is the temperature gradient of liquid (i) at the given initial state.

In a system of two-layer fluids, there are two other Rayleigh numbers and two Marangoni numbers corresponding to the upper liquid-layer and the lower liquid-layer, respectively, which are defined as follows:

$$Ra_1 = g\beta_1\Delta T_1 H_1^3/(\nu_1\kappa_1), \quad Ra_2 = g\beta_2\Delta T_2 H_2^3/(\nu_2\kappa_2);$$

$$Ma_1 = (-\partial\sigma/\partial T)\Delta T_1 H_1/(\mu_1\kappa_1), \quad Ma_2 = (-\partial\sigma/\partial T)\Delta T_2 H_2/(\mu_2\kappa_2).$$

Here ΔT_i $(i = 1, 2)$ is the local temperature difference applied across the i-th liquid.

The ratio of Rayleigh number Ra to Marangoni number Ma for a system of two-layer liquids is given by

$$\Gamma = Ra/Ma = g\beta_2\rho_2H^2/(-\partial\sigma/\partial T),$$

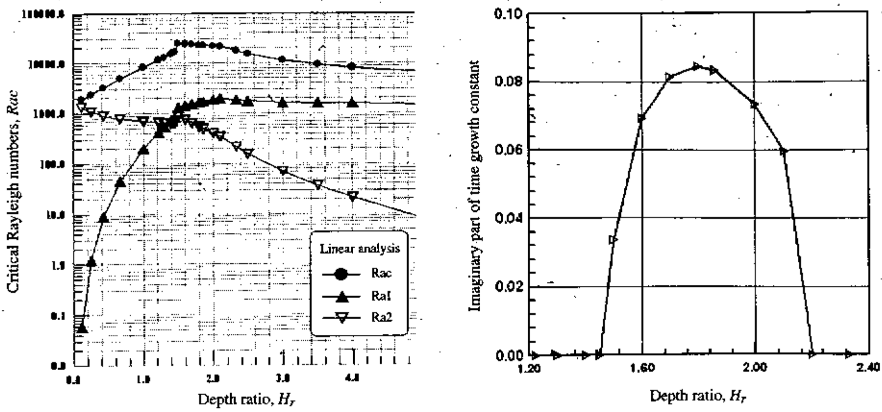
which represents the relative magnitude of thermogravitational convection and interfacial tension-driven convection and can be controlled by simply varying the total depth H of the two-layer system.

3. Linear instability results

A real two-liquid system of Silicone oil (10cSt) (in the upper layer) over Fluorinert FC70 (in the lower layer) is selected here since this pair of fluids has been more recently investigated theoretically [12] and experimentally [10]. The ratios of their physical properties are, respectively, $\kappa^* = 2.762$, $\beta^* = 1.1$, $\chi^* = 1.917$, $\mu^* = 0.344$, $\rho^* = 0.482$, $\nu^* = 0.714$, and $Pr = 406$. The interface-tension temperature coefficient of the system is taken as $\partial\sigma/\partial T = -4.46 \times 10^{-5} N/mK$.

We analysed the oscillatory instability regime of R-M-B convection in this system with a total depth $H = 6$ mm and in the ground gravity condition $g = 9.8$ ms^{-2} for a larger range of two-layer thickness ratios ($H_r = H_1/H_2$) from 0.2 to 5.0. In the case $H = 6$ mm, the corresponding ratio $\Gamma = Ra/Ma$ = 15.35 is about four times less than the value of $\Gamma = 61.38$ for the same liquid system with $H = 12$ mm, discussed in the experiments of Degen et al. [10] and analysed theoretically by Renardy and Stoltz [12]. When $H = 6$ mm, the oscillatory instability at the onset of convection is found here in the region of $1.5 < H_r < 2.1$, in which the maximum of the imaginary part of λ, $\lambda_i = 0.085$ corresponds to $H_r = 1.8$ shown in Figure 2b. When increasing H_r progressively from 1.5 to 2.1, the critical Rayleigh numbers decrease from 25010 to 21520, and in the case $H_r = 1.8$ for the critical values $Rac = 22983$ and $kc = 5.025$, the convective oscillation has the most intensity. It is notable that the transition from the monotonic onset to the oscillatory one occurs at the depth ratio $H_r = 1.5$, near the intersection ($H_r = 1.361$) of the two neutral curves of $Ra1$ and $Ra2$. Here the depth ratio $H_r = 1.361$ is the balance point ($Ra_1 = Ra_2$) of this two-layer system.

Figure 3 presents the influence of the thermocapillary effect on the instability of the system by comparing the different cases $\Gamma = 15.35$, $\Gamma = -15.35$, and $\Gamma = \infty$ ($Ma = 0$). When considering the Marangoni effect at the interface, the neutral stability curve of the system displaces to the right in comparison with the Rayleigh–Bénard instability states of the system without the Marangoni effect ($Ma = 0$) considered in Colinet and Legros's work [4]. In the case of the Rayleigh–Bénard instability of the system when neglecting the thermocapillary effect ($\Gamma = \infty$), the oscillatory onset does not exist in this



(a) Ra_0, Ra_i, and Ma_0, Ma_i (b) Corresponding critical frequency

Figure 2 Variation of the critical parameters Ra_0, Ra_i, and Ma_0, Ma_i for different depth ratios H_r and the corresponding critical frequency in a Silicone oil–Fluorinert two-layer system ($\Gamma = Ra/Ma = 15.35$).

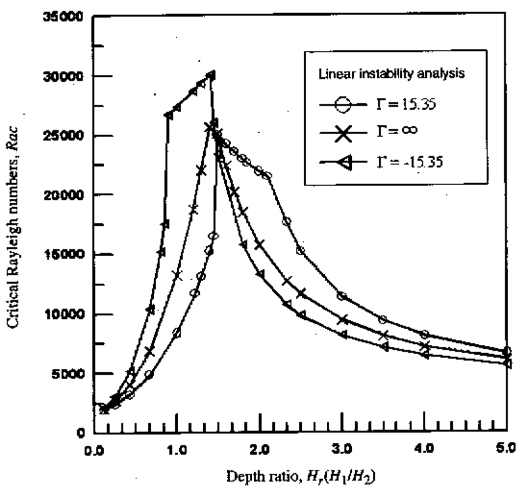


Figure 3 Comparison of the neutral stability curves for the system with different Marangoni effects: normal ($Ma > 0$), abnormal ($Ma < 0$), and without Marangoni effect ($Ma = 0$).

system (the same as the results of Renardy and Stoltz [12]). When considering the Marangoni effect at the interface, a larger oscillatory regime for $1.5 < H_r < 2.1$ is found in the R-M-B convective instability of the system. In the sense of the Marangoni effect with a positive value of $\partial\sigma/\partial T$, the oscillatory regime at the onset appears in the region of $0.892 < H_r < 1.41$ when assuming $\partial\sigma/\partial T = 4.46 \times 10^{-5} N/mK$ for $\Gamma = -15.35$.

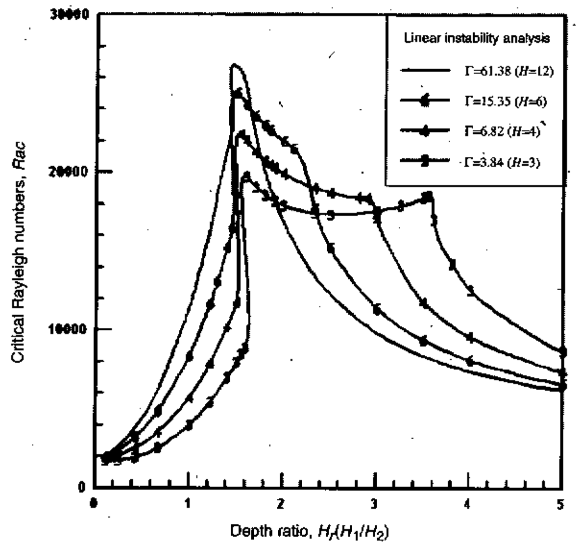


Figure 4 Oscillatory convection at the onset in the system of Silicone oil (10cSt) over Fluorinert (FC70) for different total depths H of two layers.

The oscillatory convection regions at the onset state are given in Figure 4 for different depths H of two-layer fluids. The R-M-B convective instability in the four different cases $H = 12, 6, 4$, and 3 mm has been investigated numerically. Here we consider both thermogravitational and thermocapillary effects, which may be represented by $\Gamma = Ra/Ma$.

A narrow gap $1.461 < H_r < 1.564$ of the oscillatory onset of the Rayleigh–Marangoni–Bénard convection is found in the neutral stability curve of Rac–H_r plane for $\Gamma = 61.38$ ($H = 12$ mm). The corresponding critical Rayleigh number Ra_c of the system decreases from 26840 to 26321, and the critical wave number kc falls from 5.13 to 5.08 when increasing H_r from 1.461 to 1.564. When the total depth H of the system is reduced from 6 to 3 mm, the oscillatory instability at onset occurs in the larger gap regions of the two-layer depth ratio H_r from 1.5∼2.1 to 1.6∼3.5. This variation in the gap regions is due to the augment of thermocapillary effect at the interface (represented by the decrease of the value $\Gamma = Ra/Ma$ from 15.35 to 3.84 given in Figure 4).

4. Numerical simulation

A finite volume method with SIMPLEC (Consistent Semi-Implicit Method for Pressure Linked Equations) was used for the 2D numerical simulation of the non-linear problem of R-M-B convection in the rectangular cavity with the aspect ratio of $A = L/H = 10$. The governing equations are discretised

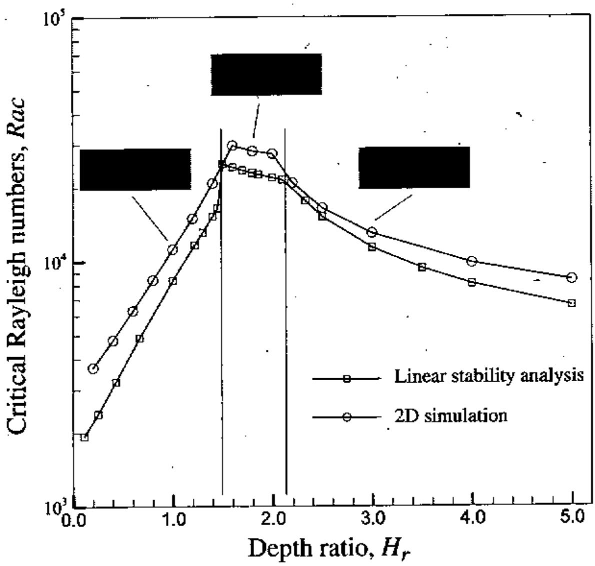


Figure 5 Critical Rayleigh numbers (linear instability analysis and 2D simulation) and the convection structures (streamlines) for different depth ratios: (right) mechanical (viscous) coupling at $H_r = 1.0$; (center) oscillatory state at $H_r = 1.8$; (left) thermal coupling at $H_r = 3$, with $\Gamma = 15.35$.

on a staggered grid using QUICK difference for convective terms and central difference for diffusive terms. An implicit three-level second-order scheme is constructed for the unsteady simulation. The calculations are carried out using a deferred correction method on a $(31 + 31) \times 501$ mesh of uniform grids. The deferred correction procedure has two-order precision and converges at approximately the rate obtained for a pure upwind approximation [15].

4.1. Critical oscillatory instability in the Silicone oil–Fluorinert liquid system

In our 2D numerical simulation of Rayleigh–Marangoni–Bénard instability in the Silicone oil (10cSt) and Fluorinert FC70 system with $H = 6$ mm, similar three types of coupling modes have been detected when the depth ratio changes within a large range from $H_r = 0.2$ to 5.0. The variation in the critical Rayleigh numbers as a function of H_r are presented in Figure 5 for both theoretical and numerical results. In the region of smaller thickness ratio $H_r = 0.2$ to 1.6, the coupling mode between the two layers is the mechanical coupling (MC), as shown typically for $H_r = 1$ in Figure 5 (left). For a larger thickness ratio, $H_r = 2.2$ to 5.0, the coupling mode is the thermal coupling (TC) and the corresponding convective structure is shown in Figure 5 (right) for $H_r = 3$. In this case, small counter-rotating sandwich cells are developed near the interface in the upper layer. While $1.6 < H_r < 2.2$, the time-dependent oscillatory convection regime appears and a constant phase offset

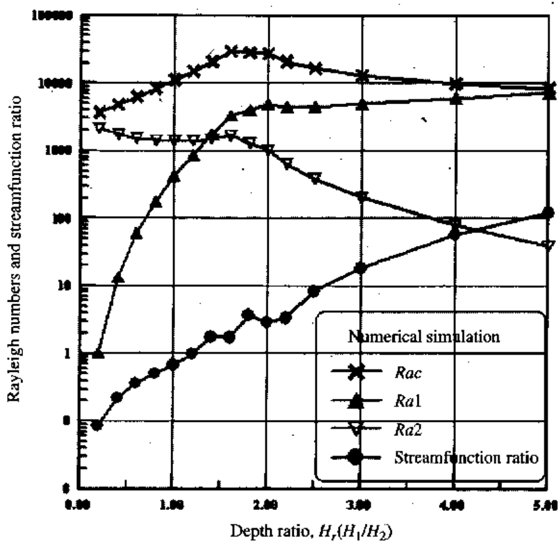


Figure 6 Critical Rayleigh numbers, Rac, Ra_1, Ra_2 of two-liquid layers with $H = 6$ mm and the streamfunction ratios of two layers for different H_r.

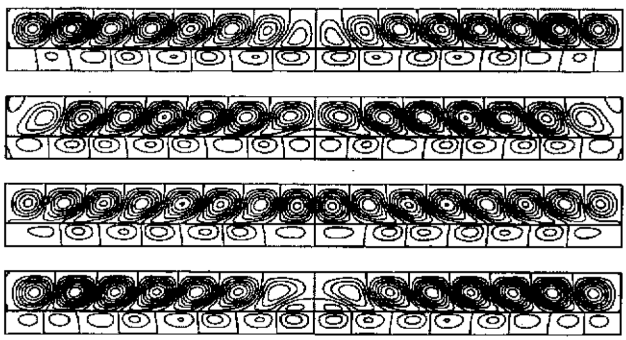


Figure 7 Time variation of the streamlines at four instants in a period of the critical oscillation flow for $H_r = 1.8$, $A = 10$, and $\Gamma = 15.35$ ($H = 6$ mm) in the Silicone oil–Fluorinert two-layer system. Time sequence is aligned downwards.

exists between the roll patterns, so that the convective mode of the system is neither TC nor MC; see Figure 5 (middle).

The critical Rayleigh numbers, Rac, Ra_1, Ra_2, and the streamfunction ratios of the two-liquid layers with $H = 6$ mm are plotted in Figure 6 for different H_r. As the prediction of the linear instability analysis, the numerical results show an intensity oscillatory convection mode when the thickness ratio is close to 1.8. In this case, the oscillatory period is 2.8 min and the dimensionless wave number is $kc = 5.02$. The corresponding streamfunction contours at four different instants within one oscillation period $P = 2.8$ min at 0, $1/4P$, $1/2P$, $3/4P$ are presented respectively in Figure 7 for $H_r = 1.8$ and

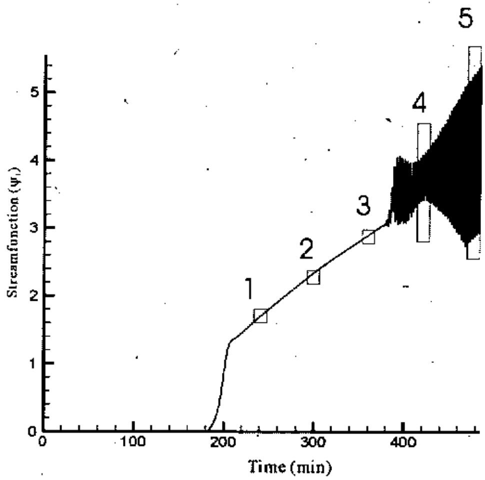
$\Delta T = 6.5°C$. The flow field remains mirror symmetric to the centre of the cavity ($x = A/2$). One can see that during one oscillation period a pair of convective rolls occurs at two lateral sides at first in each layer, and then travels continuously to the centre. It is evident that in this case the oscillatory instability mode of the system is travelling wave one. This oscillatory instability regime is a result of the competition between the MC and the TC in the two liquid layers.

4.2. Secondary oscillatory instability in the Silicone oil–water liquid system

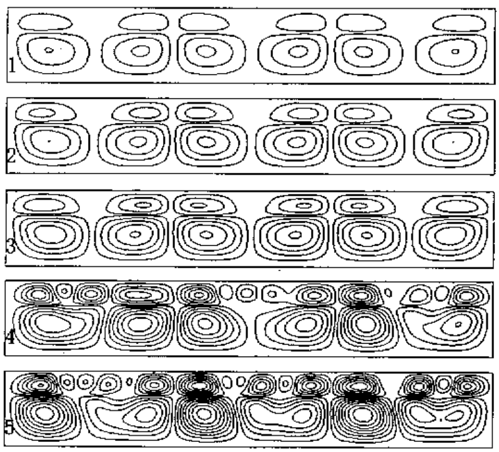
We studied another two-layer liquid system [Silicone oil (2cSt) over water] numerically. This system had been investigated experimentally [10] and theoretically [12] by other scientists. For the purpose of comparison, we used the same parameters as used in [10, 12]. However, in the linear stability analysis of [12], they did not take the Marangoni effect into consideration. We need one more parameter $\partial\sigma/\partial T$ to describe the Marangoni effect. Here we assumed $\partial\sigma/\partial T = -1.0 \times 10^{-4} N/mK$ and this is reasonable as discussed in [16]. To investigate the onset of convection, a time-consuming equivalent heating process should be applied in steps. Our strategy was to apply a faster heating process (i.e., a uniform heating rate of 0.1°C/h) to find out the bifurcation phenomenon in the two-layer system first. Then we performed unsteady calculations again for the convection mode under a fixed temperature difference to confirm that the observation we obtained is a physical one.

For the Silicone oil–water system, the main finding from our numerical simulation is that convective instability of the system will take two bifurcation processes from the static state to time-dependent convection. A typical time variation behaviour of this system is shown in Figure 8. For an initially static Silicone oil–water system, it loses its stability and onset to steady convection at $Rac(steady) = 1.1 \times 10^4$. Then it undergoes a secondary bifurcation to a time-dependent convection at $Rac(oscillation) = 2.2 \times 10^4$.

We summarised different results obtained by several authors in Table 1. Linear stability analysis predicts that the Silicone oil–water two-layer system will lose its stability and onset to steady convection, no matter whether or not we consider the Marangoni effect. Our numerical results and experimental observation agree well on the onset of oscillation. Degen et al. did not find steady convection in their experiments. This could be explained from the temperature distribution in the two-layer system. As shown in Figure 8, when the system goes into steady convection, the isotherm lines remain flat in the oil layer. "Unfortunately, using water presents another difficulty in that its variation of index of refraction with temperature is small" [10], thus a shadowgraph method would not detect the steady convection. When the convection becomes oscillatory, isotherms in the oil layer curl up and then are detected in experiments.



(a) Maximum streamfunction in lower layer

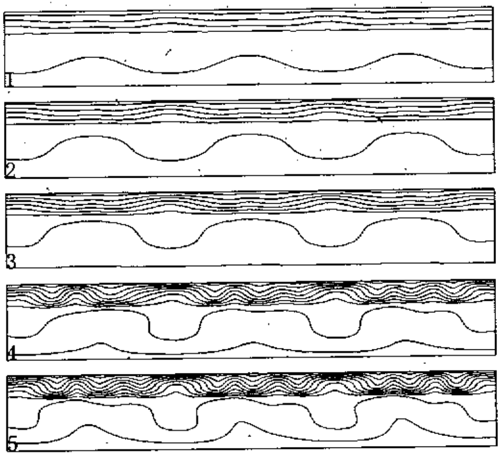


(b) Streamlines

Figure 8 Time variation of maximum streamfunction in lower layer and corresponding streamlines and isotherms at five instants in Silicone oil–water system for depth ratio $H_r = 0.5$, $A = 6.5$, and $\Gamma = 2.9$ ($H = 12$ mm).

5. Conclusion

The feature of oscillatory instability of the Rayleigh–Marangoni–Bénard convection in a thin two-layer system has been studied here by considering the real thermocapillary effect at the interface in the two-layer system Silicone oil (10cSt)–Fluorinert FC70. There exists an oscillatory convection region due to the competition between the thermocapillary forces and the buoyancy forces. The Marangoni effect enlarges the region of oscillatory regime for the layer thickness ratio H_r in the R-M-B instability in comparison with that in



(c) Isotherms

Figure 8 (continued)

Table 1 Comparison of critical *Ra* for the onset of steady (*Ra*(st)) and oscillatory convection (*Ra*(os)) in the Silicone oil–water system obtained by different methods, with $H_r = 0.5$, $A = 6.5$, and $\Gamma = 2.9$ ($H = 12$ mm).

Method	*Rac*(st)	*Rac*(os)
Experiments [10]	no	2.1×10^4
Linear stability ($Ma = 0$) [12]	1.0×10^4	no
Linear stability ($\Gamma = 2.9$)	0.70×10^4	no
2D simulation ($\Gamma = 2.9$)	1.1×10^4	2.2×10^4

the Rayleigh–Bénard instability of the system without considering the thermocapillary effect ($Ma = 0$). The oscillatory instability phenomena at the onset in the system were confirmed by our numerical simulation investigation for the non-linear instability problems. In the transition between two basically coupling modes, MT and TC, the travelling wave of the oscillatory convection at the onset of R-M-B instability is detected via the direct numerical simulation in the two-layer fluid system. This travelling wave is a result of the competition between the Rayleigh–Bénard the instability and the interfacial Marangoni effect. The typical intermediate Marangoni convection cells near the interface between two-liquid layers were observed first in the thermal coupling mode.

For the Silicone oil–water system, it is found that the two-layer system will lose stability and onset to steady convection firstly, then the steady convection bifurcates to oscillatory convection with increasing *Ra*. This secondary oscillatory instability mechanism explains the difference between the experimental observation of Degen et al. [10] and the linear stability analysis of Renardy and Stoltz [12]. Our results show the strong effects of thermocapillary force at the interface on the time-dependent oscillations at or after the onset of convection.

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