RAYLEIGH-MARANGONI-BÉNARD INSTABILITY IN TWO-LAYER FLUID SYSTEM

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ABSTRACT: Rayleigh-Marangoni-Bénard instability in a system of two-layer fluids is studied numerically. The convective instabilities in the system of Silicon Oil (10cSt) and Fluorinert (FC70) liquids have been analyzed. The critical parameters at onset of convection are presented in a large range of two-layer depth ratios from 0.2 to 5.0. Numerical results show that the instability of the two-layer system depends strongly on its depth ratio. When the depth ratio increases, the instability mode changes from mechanical coupling to thermal coupling. Between these two typical coupling modes, a time-dependent oscillation is detected. Nevertheless, traveling wave states are found in the case of oscillatory instability. The oscillation mode results from the competition between Rayleigh instability and Marangoni effect.

KEY WORDS: Rayleigh-Marangoni-Bénard convection, instability, two-layer liquids, numerical simulation

1 INTRODUCTION

Since Bénard’s (1900) experimental study of the regular hexagon convective pattern in a thin liquid layer heated from below, the onset of this “cellular” motion in an initially stationary fluid has been extensively studied theoretically and experimentally[1,2]. Two principal cellular convective mechanisms have been identified: (i) buoyancy resulting from thermally induced density gradients (Rayleigh-Bénard convection) and (ii) interfacial forces due to surface tension variations produced by temperature or concentration gradients (Marangoni convection). They have been known as Rayleigh-Bénard instability and Marangoni instability, respectively. The Rayleigh-Marangoni-Bénard instability combined with buoyancy and Marangoni effects in a single fluid layer was firstly analyzed by Nield[3]. He showed that when the two (conviction-driven) mechanisms reinforce one another they are coupled closely and also influence the stability limit.

The thermocapillary convective instabilities in a system of multi-layer liquids (two or three immiscible liquid layers) were studied by many scientists, related to several interfacial phenomena in nature such as a layered convection in the Earth’s mantle[4], and numerous engineering applications in film processing, multilayer extrusion and multilayer coating, etc. A model of two-layer liquids was constructed on the earth mantle convection with the upper layer extending to about a 700-km depth[5]. The liquid encapsulation process of crystal growth from the melt has generated recently research interest on the interfacial thermocapillary effects, particularly in space processing. The encapsulation was found to be useful for a better control of heat transfer as shown by Johnson[6], and also has the advantage of reducing or even eliminating the convective flow in the melt[7]. With respect to basic research, more attentions were focused on the instability analyses of multi-layered convection in the case of external thermal gradient perpendicular to liquid-liquid interface, for example, the classic problems of Rayleigh-Bénard convection[4,8,9] or
Marangoni-Bénard convection\textsuperscript{10–12} in two-layer liquid systems.

The simple superposition of a second convective layer on a single one brings about new elements that can produce qualitatively new behaviors\textsuperscript{[1a]}. The most important is the instability state at the convective onset of the system. The possibilities of the onset of oscillatory instability were studied quite extensively in two-layer Rayleigh-Bénard systems by instability analysis. For example, Renardy and Joseph\textsuperscript{[8,14]} conducted fairly extensive analytical studies on the stability of a two-layer Bénard system using perturbation theory. Their findings indicate that the onset of instability could be oscillatory. The linearized perturbation analysis of the system performed by Rasenat et al.\textsuperscript{[4]} revealed that oscillatory instability is possible due to a cyclic variation between viscous and thermal coupling. Colinet and Legros\textsuperscript{[9]}, using a weakly non-linear analysis, confirmed the appearance of Hopf bifurcation at the onset of convection. More recently, Degen, Colovas, and Anderer\textsuperscript{[15]}, observed experimentally a traveling wave pattern at the onset of the convective flow in the two-layer system of Silicone oil over water.

In order to understand the competition between Rayleigh-Bénard instability and Marangoni instability, and the interaction between the two liquid layers, Rayleigh-Marangoni-Bénard instability in a system of real two layered liquids have been studied in the present paper. The critical parameters are obtained by a two-dimensional unsteady numerical simulation for the two-layer system of Silicone Oil (10cSt)-Fluorinert (FC70) liquids, with the depth ratio ranging from 0.2 to 5. Calculations show that the convection in the two layers at the onset could be mechanically (viscously) coupled, thermally coupled or in the form of oscillatory instability, and the onset of oscillations is an intermediate state between the states of viscous and thermal coupling. Nevertheless, traveling waves are found in the case of oscillatory instability.

2 PHYSICAL PROBLEM AND MATHEMATICAL FORMULATION

Consider the two-layer fluid system as shown schematically in Fig.1. The dimension of the rectangular cavity is \( L \times (H_1 + H_2) \). The thickness of the upper fluid layer is \( H_1 \); the lower layer \( H_2 \), and the thickness ratio of layers is \( H_r = H_1/H_2 \). The system is heated from below with a heating rate of 1°C/h and the temperature at the bottom is uniform. The upper wall is held at a fixed temperature \( T_c \).

\[
\nabla V = 0
\]

\[
\partial V_i/\partial t + V_i \nabla V_i = -C_i^p \nabla \rho_i + C_i^e \nabla^2 V_i + C_i^\theta \partial \theta_i
\]

\[
\partial \theta_i/\partial t + V_i \nabla \theta_i = C_i^\theta \nabla^2 \theta_i
\]

where \( i = 1 \) denotes the upper-layer liquid; \( i = 2 \), the lower-layer liquid. \( V_i = (u_i, v_i, 0) \) is the dimensionless velocity; the vertical unit vector \( j \) is directed opposite to gravity; \( \theta_i = (T_i - T_c)/\Delta T \), dimensionless temperature; and \( \rho_i \), dimensionless pressure. We use \( H_2/\kappa_2, \kappa_2/H_2, H_2, \Delta T \) and \( \rho_2 \kappa_2^2/H_2^2 \) as scales for time, velocity, length, temperature, and pressure, respectively. Then the “constants” in the right hand side of these dimensionless Eqs. (1)–(3) are, respectively

\[
C_1^p = \rho_2/\rho_1 \quad C_1^e = \nu_1/\nu_2 \quad C_1^\theta = \kappa_1/\kappa_2
\]

\[
C_2^p = 1 \quad C_2^e = \beta \quad C_2^\theta = 1
\]

where \( Pr = \nu_2/\kappa_2 \) is the Prandtl number of liquid-2, \( \rho_1 \) the density, \( \nu_1 \) the kinematical viscosity, \( \alpha_i \) the expansion coefficient, \( g \) the gravitational acceleration, \( \Delta T = T_h - T_c \) the external temperature difference between the lower and upper heating walls and \( \kappa_i \) the thermal diffusivity.

On all the boundaries, the no-slip condition is satisfied. On the interface, the normal components of velocities vanish, and the continuity condition is
satisfied for temperature, heat flux, tangential components of velocities and stresses. The upper wall is maintained at a constant temperature $T_c$ and the lower wall, with temperature $T_h$, is heated from $T_c$ to a higher temperature $T_c + \Delta T$. Assume that the other side-walls are adiabatic. The surface tension is assumed to vary linearly with temperature at the interface, we have: $\sigma = \sigma_0 - \gamma(T - T_0)$, where $T_0$ is the reference temperature; and $\gamma = -\partial \sigma / \partial T$, which is the temperature coefficient of surface tension. Then the boundary conditions can be expressed as follows:

1. at the two vertical side-walls ($x = 0$ and $x = L/H_2$)

$$u_i = v_i = 0$$ (4)
$$\partial \theta_i / \partial x = 0$$ (5)

2. at the horizontal walls: ($y = -1$ and $y = H_r$)

$$u_i = v_i = 0$$ (6)
$$\theta_2 = \text{const} (y = -1) \quad \theta_1 = 0 (y = H_i)$$ (7)

where “const” is a constant which represents the heating rate. We take $c = 3,600 \kappa_2/(H_2^2 \Delta T)$ in our numerical simulation for the heating rate of $1^\circ\text{C}/\text{h}$, corresponding to the case in the experiments of Zhang et al. \[16\].

3. at the interfaces: ($y = 0$)

$$v_1 = u_2$$ (8)
$$v_1 = v_2 = 0$$ (9)
$$\partial u_2 / \partial y - \frac{\mu_1}{\mu_2} \partial v_1 / \partial y = -M a \partial \theta_2 / \partial x$$ (10)
$$\frac{\lambda_1}{\lambda_2} \partial \theta_1 / \partial y = \partial \theta_2 / \partial y$$ (11)
$$\theta_1 = \theta_2$$ (12)

Equations (8)~(12) represent the continuity of tangential velocity, kinematical condition, the balance of tangential stresses, the continuity of heat flux, and the continuity of temperature, respectively. Here $\lambda_i$ is the thermal conductivity. Marangoni number is defined as $Ma = \gamma \Delta T H_2 / \mu_2 \kappa_2$.

The initial temperature field is uniform and the fluids are at rest before the onset of Rayleigh-Marangoni-Benard convection in the system.

In a system of two-layer fluids, three Rayleigh numbers and three Marangoni numbers should be identified corresponding to the total system of two layers, the upper layer and the lower layer, respectively, in order to understand the coupling mechanisms between the upper and lower layer fluids. They are the total Rayleigh number calculated using parameters of fluid 2, $Ra_0 = g \rho_2 \alpha (H_1 + H_2)^3 / \nu_2 \kappa_2$; and the Rayleigh number of each liquid layer, $Ra_i = g \alpha_i \Delta T_i H_i^3 / \nu_i \kappa_i$.

Similarly, the total Marangoni number is defined as $Ma_0 = \gamma \Delta T (H_1 + H_2) / \mu_2 \kappa_2$, and the Marangoni numbers for fluid 1 and fluid 2

$$Ma_i = \gamma \Delta T_i H_i / \mu_i \kappa_i$$

where $\Delta T_i$ is temperature difference of each layer at the rest state. As detailed by Degen et al.\[15\], $\Delta T_i$ is calculated using the so-called “conduction assumption”. Use of this assumption gives an interface temperature of

$$T_{int} = \frac{\lambda_1 H_2 T_c + \lambda_2 H_1 T_h}{\lambda_1 H_2 + \lambda_2 H_1}$$

and then $\Delta T_1 = T_{int} - T_c$, $\Delta T_2 = T_h - T_{int}$, respectively.

### 3 NUMERICAL METHOD

Numerical simulation is carried out by a finite difference method in two dimensions. SIMPLEC method, Consistent Semi-Implicit Method for Pressure Linked Equations, is used in the solution of the non-linear partial differential equations. Here the equations are discretized on a staggered grid using upwind difference for the convective term and central difference for diffusive terms.

In this paper, the calculations are carried out using a $(31 + 31) \times 501$ mesh of uniform grids. The grid-dependence of the solution was checked by comparing solutions obtained for different grids from $(31 + 31) \times 251$ mesh to $(61 + 61) \times 501$ mesh. For example, we get the critical Rayleigh number $Ra_c = 1.108 \times 10^4$ for a $(31 + 31) \times 251$ mesh, and $Ra_c = 1.180 \times 10^4$ for a $(61 + 61) \times 501$ mesh.

The validation of the numerical code is established by comparing our results with experimental results (critical temperature) of Zhang\[16\]. A good agreement is obtained for the convective instability in the system of Silicon Oil (10cSt) and Fluorinert (FC70) for different thickness of two layers. For example, the critical temperature $\Delta T_c$ observed experimentally for $H_1 = 1.210$ mm (Silicon Oil (10cSt) layer) and $H_2 = 4.972$ mm (Fluorinert (FC70) layer) is 0.89°C, and the numerical one giving by our code is 0.84°C. Another situation is 4.161 mm thick of Silicon Oil over 1.477 mm of Fluorinert liquid, the critical
temperature observed is 4.56°C from experiment and 4.23°C as obtained from numerical calculation.

Figure 2 shows the variation of the maximum non-dimensional stream function $\psi_{\text{max}}$ as a function of time. For a linear variation of temperature difference with time, it means that below a temperature difference ($\Delta T_c$) the value of $\psi_{\text{max}}$ is nearly 0, which indicates that the fluid is still at rest. When the temperature difference ($\Delta T$) continues increasing very slightly, the value of $\psi_{\text{max}}$ gets a rapid increase, which corresponds the state where the convective flow is formed step by step in the system of two-layer liquids. In the present paper, the critical parameters corresponding to the time at point “c” for the onset of Rayleigh-Marangoni-Bénard convection are determined numerically from the nonlinear development of $\psi_{\text{max}}$ as shown in Fig.2.

![Figure 2](image)

**Fig.2** Time variation of the maximum non-dimensional steam function for depth ratio $H_r = 1$

4 RESULTS

4.1 Onset of the Convection

Many combinations of liquids are possible to form an immiscible liquid system. The Rayleigh-Marangoni-Bénard instabilities in a real two-layer liquid system of Silicon Oil (10cSt) and Fluorinert (FC70) liquids are studied numerically in this paper. These two immiscible liquids were used extensively for the experimental investigations on the ground or in microgravity environment\[13,16,17\]. Their physical properties are listed in Table 1. Since the interfacial tension of Silicone 10cSt-Fluorinert FC70 is about $7.6 \times 10^{-3}$ N/m, the Crispation number $Cr = \mu \sigma / d$ becomes $O[10^{-5}]$ if the layer depth is in the order of millimeter. This small value of Crispation number indicates that the fluid system may justify the assumption of non-deformable interface in the present analysis.

Calculations are completed for depth ratio varying from 0.2 to 5.0 in order to show the complicated and colorful instability behaviors of two-layer fluid convection. Critical temperature difference and critical wave number $k_c = 2\pi (H_1 + H_2)/\lambda_c$, where $\lambda_c$ is the wavelength at the onset of Rayleigh-Marangoni-Bénard convection, are listed in Table 2. With a fixed total depth of two layers $H_1 + H_2 = 6$ mm, and $L = 60$ mm, the critical temperature differences $\Delta T_c$ changes from 0.85°C for $H_r = 0.2$°C to 6.84°C for $H_r = 1.6$.

The critical parameters at the onset of Rayleigh-Marangoni-Bénard convection in the system are shown in Fig.3 for different depth ratios $H_r$. The neutral curve of the total Rayleigh number for the system indicates that the system is the most stable at $H_r = 1.6$ where the Silicone Oil 10cSt layer is 1.6 times as deep as the FC70 layer. The corresponding critical Rayleigh numbers are $Ra_{oc} = 29730$, $Ra_{1c} = 3314$ and $Ra_{2c} = 1692$, respectively for the total depth, upper layer depth and lower layer depth. In contrast to $H_r = 1$, at the most stable point, $Ra_1$ is greater than $Ra_2$ and $Ma_1$ greater than $Ma_2$. The total Rayleigh number increases from 3693 at $H_r = 0.2$ to 29730 at

<table>
<thead>
<tr>
<th>$H_r(H_1/H_2)$</th>
<th>0.2</th>
<th>0.4</th>
<th>0.6</th>
<th>0.8</th>
<th>1.0</th>
<th>1.2</th>
<th>1.4</th>
<th>1.6</th>
<th>1.8</th>
<th>2.0</th>
<th>3.0</th>
<th>4.0</th>
<th>5.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta T_c/°C$</td>
<td>0.85</td>
<td>1.10</td>
<td>1.45</td>
<td>1.93</td>
<td>2.58</td>
<td>3.45</td>
<td>4.80</td>
<td>6.84</td>
<td>6.50</td>
<td>6.33</td>
<td>3.00</td>
<td>2.27</td>
<td>1.93</td>
</tr>
<tr>
<td>$k_c$</td>
<td>3.14</td>
<td>3.14</td>
<td>3.77</td>
<td>3.77</td>
<td>4.40</td>
<td>4.40</td>
<td>5.65</td>
<td>7.54</td>
<td>5.02</td>
<td>5.02</td>
<td>3.77</td>
<td>3.14</td>
<td>3.14</td>
</tr>
</tbody>
</table>

Table 1 Physical properties of the liquids

<table>
<thead>
<tr>
<th>$\rho/(kg.m^{-3})$</th>
<th>$\mu/(kg.m^{-1}.s^{-1})$</th>
<th>$\kappa/(m^{2}.s^{-1})$</th>
<th>$\lambda/(J.m^{-1}.s^{-1}.K^{-1})$</th>
<th>$\alpha/K^{-1}$</th>
<th>$\gamma/(N.m^{-1}.K)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicone 10cSt</td>
<td>$9.35 \times 10^2$</td>
<td>$9.35 \times 10^{-3}$</td>
<td>$9.5 \times 10^{-8}$</td>
<td>0.134</td>
<td>$1.1 \times 10^{-3}$</td>
</tr>
<tr>
<td>FC70</td>
<td>$1.94 \times 10^3$</td>
<td>$2.72 \times 10^{-2}$</td>
<td>$3.4 \times 10^{-8}$</td>
<td>$6.99 \times 10^{-2}$</td>
<td>$1.0 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

Table 2 Critical temperature difference and critical wave number $k_c = 2\pi (H_1 + H_2)/\lambda_c$ for the system of Silicone 10cSt-Fluorinert FC70 liquids (heated from below)
4.2 Coupling Modes of the Two-layer System

To determine the primary instabilities as a function of the depth ratio of two fluids is the main topic of the present study. The depth ratio is a vital parameter with respect to the onset of convective instability in a system of two-layer fluids, but little attention has been paid to that in previous investigations. Colinet and Legros\cite{9} presented a linear and weakly nonlinear analysis on the Hopf bifurcation of the two-layer Rayleigh-Bénard convective instability in a model system, where the parameters taken for analysis are $\kappa_1/\kappa_2 = 0.5$, $\mu_1/\mu_2 = \lambda_1/\lambda_2 = 1$, and $\nu_1\alpha_2/\nu_2\alpha_1 = 2$ and the Marangoni effect (instability) at the interface was disregarded. In this special case, they predicted that for small upper layer depth when $H_r < 0.92$, the roll pattern in the two-layer system should be mechanically (viscously) coupled (MC, i.e. rising fluid over falling fluid, and vice versa, so that superposed rolls rotate in opposite directions). For a thick upper layer $H_r > 1.067$, the pattern should be thermal coupled (TC, i.e. rising plumes over rising plumes, with falling fluid aligned with falling fluid). In the range of two-layer depth ratios $H_r = 0.92$ to 1.067, one expects a competition between MC and TC modes.

In our 2D numerical simulations of Rayleigh-Marangoni-Bénard instability in a real two-layer liquid system, similar three types of coupling modes have been detected when the depth ratio changes in a large range from depth ratio $H_r = 0.2$ to 5.0. For smaller depth ratios from 0.2 to 1.4, the coupling mode between the two layers is MC, as shown in Fig.5(a). For larger depth ratios $H_r = 2.5 \sim 5.0$, the coupling mode is TC (Fig.5(b)). In this case, small counter-rotating sandwich-cells develop near the interface in the upper layer. But this does not change the fact that there are two basically different types of coupling between superimposed layers. While $1.4 < H_r < 2.5$, a constant phase offset exists between the roll patterns, so that the system is neither TC nor MC (Fig.5(c)). This is a traveling pattern as detailed below.
4.3 Oscillatory Instability

The present combination of two superposed immiscible liquid layers may result in oscillatory instability regimes. Numerical results show that the convective instability in the two-layer fluid system of Silicone 10cSt over Fluorinert FC70 is of an oscillatory mode when the depth ratio is close to 1.8, from 1.6 to 2.2. Figure 6 shows the development of the maximum stream functions in both layers near the critical point for \( H_r = 1.8 \). The amplitude of the oscillation for the maximum stream function is about 30% of the average maximum stream function over time in each layer. The oscillatory period is 2.8 min.

Stream function contours and isotherms are shown in Fig.6 for \( H_r = 1.8 \) at a smaller distance from the onset of convection. The flow field remains mirror symmetric to the vertical line at the center of the cavity \( (x = L/2) \) and only the right half part of full convective fields is presented here. Four instants within one oscillation period \( P \) at \( 0, 1/4P, 1/2P, 3/4P \) are shown in Fig.7. One can see that during
one oscillation period \( P = 2.8 \text{ min} \) a pair of convection rolls occurs at the left side in each layer firstly, and then travels continuously to the right side. Correspondingly, at the right side, a pair of rolls disappears during one period. From the isotherms in Fig.6, it is clear that in this case the instability mode of the system is a traveling wave one.

Marangoni effect enhances the instability of the lower layer when the two-layer system is heated from below. When \( H_r \) is small \((H_2 > H_1)\) and \( Ra_2 > Ra_1 \), the fluid in the lower layer, after the onset to convection at first, sees mechanical coupling through the continuity of tangential velocity at the interface. When \( H_r \) is large enough, the upper layer loses stability at first due to the fact that \( Ra_1 \gg Ra_2 \), and the convection in upper layer causes the non-uniform temperature distribution along the interface. Because of Marangoni effect, a small third convection roll forms near the interface. In this case thermal coupling appears. When \( H_r \) is in the intermediate region, the upper layer competes with the lower layer. Under the interaction of Rayleigh instability and Marangoni effect, a phase shift occurs between convection rolls in both layers. The convection mode is neither mechanical coupling nor thermal coupling; both basic coupling states coexist along the interface. In this situation, Rayleigh instability and Marangoni effect cooperate to create a uniform traveling movement of the velocity and temperature field.

5 CONCLUSION

Numerical simulations of Rayleigh-Marangoni-Bénard (RMB) convection instability in a system of two-layer fluids are performed by solving the Navier-Stokes equations and energy equation. The different typical instability modes, such as MC, TC and the transition between them, in a real two-layer system of Silicone 10cSt and Fluorinert FC70 liquids are obtained numerically in the nonlinear regime, while considering the Marangoni effect induced by surface tension gradient at the interface. The nonlinear instability results of such a system are obtained in a larger range of the depth ratio of two liquid layers from 0.2 to 5.0. Numerical results indicate that the instability depends strongly on the depth ratio of two liquid layer. When increasing the depth ratio, the instability mode changes from mechanical coupling to thermal coupling. In the transition between these two basic coupling modes, the traveling wave mode corresponding to the oscillatory instability at the onset of RMB convection is detected via the direct numerical simulation in the two-layer fluid system. The traveling wave is different from previous studies of both stability analysis and experiments in Ref.[13]. The origin of this phenomenon is the competition between Rayleigh instability and Marangoni effect in multiple liquid layers.

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