Steady Marangoni instabilities in variable-viscosity liquid layer in the presence of insoluble surfactant

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Abstract. The ability to control the complex convective flow patterns is important and one of the factors that alter the dynamics of the surface tension is surface-active agents or surfactants. Surfactants affect the surface tension at the free upper surface by producing tangential stress. This study theoretically examines the onset of steady instabilities in a thin nondeformable and deformable variable-viscosity liquid layer in the presence of insoluble surfactant. The surface tension at the free surface is assumed to be linearly dependent on temperature and concentration gradients. The dynamic viscosity is assumed to be exponentially dependent on temperature. The stability problems are governed by the systems of nonlinear partial differential equations consisting of continuity, momentum and energy equations. Linear stability analysis consisting of scaling, perturbation of infinitesimal disturbances and superposition in normal modes are used to transform the system of partial differential equations into system of ordinary differential equations. The exact analytical solutions are derived and the marginal curves are illustrated to show effects of some controlling parameters particularly the variable viscosity and elasticity parameter. Viscosity variation and elasticity parameter of surfactant act as destabilizer while the Biot number and Lewis number stabilize. For deformable surface, convection sets in at long wavelength for liquid with variable viscosity but convection occurs at short wavelength for constant viscosity liquid.

1. Introduction
Convective instabilities are natural phenomena observed in nature and technological processes. In many engineering and technical processes, the ability to control either by delaying or promoting the convective motion is important in order to achieve the desired results. The two main driving mechanisms of convective instabilities in a liquid layer are buoyancy forces and surface tension forces but in a very thin layer of liquid, the surface tension forces are dominant [1]. The convective motion can also be altered by the liquid properties which can be dependent on its thermodynamic state. Strong viscosity variation and surface-active agents are known to alter the dynamic state of the convection in a liquid layer. The cosine law [2], linear [3] exponential and super exponential [4-10] viscosity variations have been examined on the stability of steady and oscillatory convections. For exponential temperature-dependent viscosity, viscosity variation destabilizes [7]. In strong viscous liquids, viscosity and surface deformability have stabilizing effects on the onset of convective instabilities [8]. Surface-active agent or surfactant has an ability to decrease the excess surface energy by exerting additional tangential stress at the interface. The stabilizing effect of surfactant on the steady convection is due to the opposite tangential forces of temperature (thermocapillary) and surfactant distribution.
(soluto-capillary) but the oscillation of temperature and surfactant disturbances induce oscillatory convection and weaken the stabilizing effect of surfactant [11]. In the presence of capillary-gravity waves in deep layer, surfactant provides weak stabilization [12, 13].

In this study, the Marangoni instability in a variable-viscosity liquid layer in the presence of insoluble surfactant is studied. The linear stability theory is applied to obtain a system of ordinary differential equations of eigenvalue problem in form of the normal modes. The system is solved analytically for closed-form solutions and the marginal curves are plotted to assess the influence of the physical parameters on the onset of steady convection for both nondeformable and deformable surface.

2. Problem Formulation

Consider a horizontal liquid layer of thickness $d$ bounded below at $z = 0$ by a rigid plate and the upper boundary at $z = d$. The liquid layer is heated from below and cooled from above. In view of the rotational symmetry, the coordinates $x$ and $z$ are considered for the problem where $x$ axis is coincidental with the horizontal plane that lies on the lower solid boundary and $z$ be the vertical axis. The two-dimensional space is sufficient for the development of disturbances for the linear stability analysis [11]. The free surface is considered deformable and the deflection of the free surface from its original position is $z = d + \xi(x, t)$ where $\xi(x, t)$ is the deflection and $t$ is the time. The liquid is assumed incompressible and the viscosity $\mu$ is assumed to be exponentially dependent on the temperature $T$ given by

$$\mu = \mu_0 \exp[-\gamma (T - T_0)],$$

where $\mu_0$ is a reference viscosity corresponding to the reference temperature $T_0$ at the free surface and $\gamma$ is a positive constant. The surface tension $\sigma$ is assumed to depend linearly on temperature $T$ and surface concentration $\Gamma$,

$$\sigma = \sigma_0 - \sigma_1(T - T_0) - \sigma_2(\Gamma - \Gamma_0),$$

where $\sigma_0$ and $\Gamma_0$ are the reference values of surface tension and concentration, respectively, and $\sigma_1$ and $\sigma_2$ are rates of change of the surface tension with respect to temperature and concentration, respectively. For a very thin liquid layer or under microgravity conditions, the buoyancy effect is negligible. Other physical properties of the liquid such as density and pressure are assumed constants. When the liquid is at rest, the hydrodynamic pressure $p_b$ and the temperature of the liquid are

$$p_b = p_a + \rho g (d - z)$$

$$T_b = T_a - \beta z$$

where the atmospheric pressure is $p_a$, gravitational acceleration $g$, $\rho$ is the density and $-\beta$ is the temperature gradient.

The system of variable viscosity liquid layer is governed by the equations of conservation of mass, momentum and energy

$$\nabla \cdot \mathbf{v} = 0$$

$$\rho \left[ \frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} \right] = -\nabla p + \nabla \cdot (2\mu \mathbf{D})$$

$$\frac{\partial T}{\partial t} + (\mathbf{v} \cdot \nabla)T = \kappa \nabla^2 T$$

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where \( \mathbf{v} = (u, w) \) is the liquid velocity in the \( x \) and \( z \) directions, respectively, \( \kappa \) is the thermal diffusivity, \( p \) is the pressure, \( \mathbf{D} \) is the deformation rate tensor.

The boundary conditions at the lower boundary \( z = 0 \) are of no-slip and uniform temperature,
\[
  u = w = 0, \quad T = T_w, \tag{8}
\]
respectively. At the upper nondeformable free surface \( z = d \), the kinematic condition and the heat transfer governed by the Newton’s law of cooling are
\[
  \lambda \frac{\partial T}{\partial n} + h(T - T_o) = 0, \tag{10}
\]
and the normal and tangential stresses balances are
\[
  \hat{n} \cdot \tau \cdot \hat{n} = 2H\sigma \cdot \hat{n} \tag{11}
\]
\[
  \tau \cdot \hat{n} = \nabla \sigma \tag{12}
\]
where \( H \) is the mean curvature, \( \tau \) is the stress tensor, \( \hat{n} \) is a unit normal vector to the upper deformable surface.

The surfactant concentration \( \Gamma \) at the free surface is considered insoluble and only localized at the surface given by [10],
\[
  \frac{\partial \Gamma}{\partial t} + \nabla \cdot (u_n \Gamma) + \epsilon w_n \Gamma = D_0 \nabla_s^2 \Gamma, \tag{13}
\]
where \( u_n \) and \( w_n \) are the tangential and normal velocities on the surface, \( \lambda \) is the thermal conductivity, \( \nabla_s \) is the surface gradient, \( h \) is the rate of heat transfer by convection, \( \hat{n} \) is the normal vector, \( \epsilon \) is the local curvature and \( D_0 \) is the surfactant diffusivity.

2.1. Linear Stability Theory
In the linear stability theory, the equilibrium state of the system (1) – (13) is perturbed with infinitesimal disturbances, nondimensionalized and linearized. The disturbances for the velocity, pressure, temperature and concentration are
\[
  \mathbf{v} = 0 + \mathbf{v}(x, z, t) = (u', w'), \tag{14}
\]
\[
  p = p_0 + p'(x, z, t), \tag{15}
\]
\[
  T = T_o - \beta z + T'(x, z, t), \tag{16}
\]
\[
  \Gamma = \Gamma_0 + \Gamma'(x, t) \tag{17}
\]
respectively.

The scaling quantities for the length, time, velocity, temperature, concentration and pressure are \( d, d^2 / \kappa, \kappa/d, \beta d, T_0 d \) and \( \mu, \kappa / d^2 \). The resulting linearized system of (5) – (7)
\[
  \frac{1}{Pr} \frac{\partial}{\partial t} \nabla^2 w = \nabla^2 \nabla^2 w + 2 \frac{df}{dz} \frac{\partial}{\partial z} \nabla^2 w + \frac{d^2 f}{dz^2} \left( \frac{2}{\partial^2 \nabla^2 w} - \nabla^2 w \right) \tag{18}
\]
\[ \nabla^2 p = 2 \frac{df}{dz} \nabla^2 w + 2 \frac{d^2 f}{dz^2} \frac{\partial w}{\partial z} \quad (19) \]
\[ \frac{\partial T}{\partial t} - \nabla^2 T = w \quad (20) \]

with velocity and temperature conditions (8) at \( z = 0 \),
\[ w = 0, \quad \frac{\partial w}{\partial z} = 0, \quad T = 0, \quad (21) \]

and at \( z = 1 \), for kinematic condition and heat flux balance (9) are
\[ w = \frac{\partial \xi}{\partial t}, \quad \lambda \frac{\partial T}{\partial z} + h(T - \xi) = 0 \quad (22) \]

respectively, for the tangential and normal balances (11) and (11)
\[ f \left( \frac{\partial^2 w}{\partial x^2} - \frac{\partial^2 w}{\partial z^2} \right) = -Ma \frac{\partial^2}{\partial x^2} (T - \xi) - N \frac{\partial^2 \Gamma}{\partial x^2} \quad (23) \]
\[ \frac{1}{Pr} \frac{\partial^2 w}{\partial z \partial t} = f \left( 3 \frac{\partial^2 w}{\partial z \partial x^2} + \frac{\partial^3 w}{\partial z^3} \right) + \frac{Bo}{Cr} \frac{\partial^2 \xi}{\partial x^2} - \frac{1}{Cr} \frac{\partial^4 \xi}{\partial x^4} + \frac{d f}{dz} \left( \frac{\partial^2 w}{\partial z^2} - \frac{\partial^2 w}{\partial x^2} \right) \quad (24) \]

and for the surfactant concentration (13)
\[ \frac{\partial \Gamma}{\partial t} - \frac{\partial w}{\partial z} = Le \frac{\partial^2 \Gamma}{\partial x^2} \quad (25) \]

where the viscosity variation function is \( f(z) = \exp[V(z - 1)] \), \( Pr \) is the Prandtl number, \( Bi \) is the Biot number, \( V \) is the viscosity parameter, \( Bo \) is the Bond number, \( Cr \) is the Crispation number and \( Le \) is the Lewis number.

The analysis of normal modes in terms of two-dimensional periodic waves is made to seek solutions of the form of the superposition of normal modes given by
\[ (w, T, \Gamma, \eta) = [W(z), \theta(z), \phi(z), Z] \exp[i(k + \alpha x)], \quad (26) \]

where \( W(z), \theta(z), \phi(z) \) and \( Z \) are the velocity, temperature, concentration and surface deflection amplitudes, respectively, \( k \) is the wave number and \( \alpha \) is the complex growth rate.

The linearized dimensionless momentum, and energy equations (18) – (20) reduce to
\[ f(z) \left( (D^2 - k^2)^2 + V^2 (D^2 - k^2) + 2V^2 k^2 \right) W = \frac{1}{Pr} \omega (D^2 - k^2) W, \quad (27) \]
\[ \left[ \omega - (D^2 - k^2) \right] \theta = -W \quad (28) \]

and the boundary conditions (21) – (25) are transformed to
\[ w = DW = 0, \quad \theta = 0, \quad (29) \]
at \( z = 1 \):
\[
W + \omega Z = 0,
\]
\[
D\theta + Bi(\theta - Z) = 0
\]
\[
f(1)\left(D^2 + k^2\right) - k^2[M(\theta - Z) + N\phi] = 0
\]
\[
f(1)\left[D^2 - 3k^2\right]DW + V\left(D^2 + k^2\right)W + \frac{k^2(Bo + k^2)Z}{Cr} = \frac{1}{Pr}\omega DW.
\]
\[
\omega \phi - DW = -k^2 Le \phi
\]

Eliminating \( \phi \) in (31) using (33), condition (31) becomes
\[
f(1)\left(D^2 + k^2\right)W - k^2\left[M(\theta - Z) + \frac{NDW}{\omega + k^2 Le}\right] = 0
\]

The dimensionless parameters are Biot number \( Bi = \frac{hH}{\lambda} \), Marangoni number \( M = \frac{\sigma_i \beta H^2}{(\kappa \lambda_i)} \), elasticity number \( N = \frac{\sigma_2 \gamma_0 H^2}{(\mu_0 \kappa)} \), Prandtl number \( Pr = \frac{hH}{\lambda} \), viscosity parameter \( V = \frac{\gamma \beta H}{h} \) and Lewis number \( Le = \frac{D_0}{\kappa} \), Crispation number \( Cr = \frac{\mu_0 \kappa}{(\sigma_0 d)} \), Bond number \( Bo = \frac{\rho gd}{\sigma_0} \).

3. **Analytical Solutions of Steady Convection**

The onset of convective instabilities is determined by the Marangoni number \( M = M_1 + iM_2 \) and the complex growth rate \( \omega = \omega_r + i\omega_i \). If \( \omega_i < 0 \) then all disturbances decay and the system is stable and if \( \omega_i > 0 \) then all disturbances grow and the system is unstable. When \( \omega = 0 \), the stability is at the marginal state in which the disturbances is neither damped nor amplified. The convection sets in as stationary convection when \( \omega_i = 0 \) and oscillatory convection when \( \omega_i \neq 0 \). Marginal and onset of convective instabilities can be analyzed for the steady convection \( \omega = 0 \).

In this study, the complex growth rate \( \omega = 0 \) is considered for the onset of steady convection. The system (27) – (32) and (34) reduces to system of ordinary differential equations with coefficients. The closed form analytical solutions can be derived for \( W(z) \), \( \theta(z) \) and the Marangoni number \( M \). The solution for \( W(z) \) that satisfies the velocity conditions in (29) and (30) is
\[
W(z) = A_1\left[\exp(c_1z) - \exp(c_2z)\right]\cos(c_3z) - [A_2\exp(c_1z) - A_3\exp(c_2z)]\sin(c_3z)
\]
where \( A_1 \) is an arbitrary constant and \( c_1 \), \( c_2 \), \( c_3 \), \( c \), \( A_2 \) and \( A_3 \) are
\[
c_1 = -\frac{V}{2} + \frac{1}{\sqrt{2}}\left(c^2 + k^2 - \frac{V^2}{4}\right)^{1/2}, \quad c_2 = -\frac{V}{2} - \frac{1}{\sqrt{2}}\left(c^2 + k^2 + \frac{V^2}{4}\right)^{1/2}
\]
\[
c_3 = 1 + k^2 - \frac{V^2}{4}
\]
\[
c = \frac{\left(k^2 + \frac{V^2}{4}\right)^{1/4} + k^2 V^{1/4}}{\frac{V^2}{4} - k^2}
\]
\[
A_2 = \cot c_3 + \frac{(c_2 - c_1)\exp(c_2 - c_1)}{c_3\left[1 - \exp(c_2 - c_1)\right]}, \quad A_3 = \cot c_3 + \frac{(c_2 - c_1)}{c_3\left[1 - \exp(c_2 - c_1)\right]}
\]

Substituting \( W(z) \) in (28), the complete solution for \( \theta(z) \) with \( \theta_p(z) \) as the particular solution corresponding to nonhomogeneous equation involving \( W(z) \) is
\[
\theta(z) = B_1 \sinh(kz) + B_2 \cosh(kz) + \theta_p(z)
\]
\[
\theta_p(z) = A_4\left[C_1 \exp(c_1z) + C_2 \exp(c_2z)\right]\cos(c_3z) + [C_3 \exp(c_1z) + C_4 \exp(c_2z)]\sin(c_3z)
\]
The closed form solutions can be obtained for the unknown variables of the surface deflection $Z$ and the Marangoni number $M$ from the conditions (30), (33) and (34).

4. Results and Discussion

The system (27)–(31), (33) and (35) reduces to the problems of [6] for variable-viscosity liquid without insoluble surfactant ($N=0$) and of [11] for constant viscosity liquid ($V=0$) with insoluble surfactant. Setting both $N=0$ and $V=0$ gives the classical Marangoni problem of [1]. The marginal curves are plotted for the effects of the viscosity variation $V$, elasticity parameter $N$, Lewis number $Le$, Biot number $Bi$, Crispation number $Cr$ and Bond number $Bo$ on the Marangoni number $M$ for the onset of steady convection. The Marangoni number $M$ measures the ratio of thermocapillary forces to viscous forces, $V$ measures the variation between the viscosities at the lower and upper surfaces, $N$ measures the surfactant’s ability to modify the surface tension at the free surface. Therefore, the considered problem involves both thermocapillary and solutocapillary convection. The Biot number measures the heat transfer at the free surface. $Bi=0$ corresponds to the situation where no heat is allowed to escape and the heat is communicated within the system and when $Bi$ increases, more heat is allowed to escape from the liquid to the upper gas phase. The Lewis number $Le$ measures the ratio of diffusive force and viscous force. The Crispation number $Cr$ measures the ratio of between viscous and surface tension forces and the Bond number $Bo$ measures the ratio of gravity waves and surface tension forces.

The marginal curves for the Marangoni number $M$ are plotted versus the wavenumber $k$ and the effects of varying parameters on the onset of steady convection are determined by the increasing or decreasing of $M$. Figs. 1 – 4 illustrate the effects of elasticity parameter $N$, viscosity parameter $V$, Biot number $Bi$ and Lewis number $Le$ for nondeformable surface.

![Fig. 1. Marginal stability curves for various $N$ when $V = Bi = Le = 0.1$.](image)

In Fig. 1, the critical Marangoni number $M_c$, indicated by the minimum value of $M$ of the marginal curves, decreases as the elasticity parameter $N$ increases. The positive value of $N$ corresponds to the existence of insoluble surfactant and the surfactant reduces the interfacial surface tension and hence promotes convective instabilities.
Fig. 2. Marginal stability curves for various viscosity variation parameter $V$ when $N = 0.03$ and $Bi = Le = 0.1$.

Fig. 3. Marginal stability curves for various Biot number $Bi$ when $N = 0.03$ and $V = Le = 0.1$.

In Fig. 2, the minimum value of the Marangoni number $M$ decreases as the viscosity variation $V$ increases. Higher difference in viscosity between the lower and upper surface enhances the heat transfer and thus induces the onset of convective motion to destabilize the liquid system. The minimum value of $M$ is shown to increase in Fig. 3 as the Biot number $Bi$ increases. The Biot number $Bi$ characterizes the heat transfer at the upper free surface and more heat is allowed to escape from the system leads to a more stable liquid layer.

Fig. 4. Marginal stability curves for various Lewis number $Le$ when $N = 0.03$ and $V = Bi = 0.1$.
Fig. 4 illustrates the effect of Lewis number on the onset of steady convection. There is small variation in the effects of Lewis number as it approaches 1. The Lewis number is close to unity indicates that thermal boundary layer and mass transfer by diffusion are comparable and the system remains stable. For the Lewis number below than 1, the stability depends on the destabilizing effect of the temperature.

![Fig. 4. Marginal stability curves for $Bi = Le = Bo = 0.1$, $Cr = 0.001$ and deformable surface.](image)

The marginal curves for the steady Marangoni convection for deformable surface with constant viscosity ($V = 0$) and variable viscosity ($V = 0.1$) in the presence of surfactant is on the stability of the liquid layer. In the absence of viscosity variation, the steady convection sets in at the short wavelength but viscosity variation results in the steady convection at long wavelength. Viscosity variation is a destabilizing factor.

5. Conclusion
The effects of viscosity variation and the insoluble surfactant on the instabilities of the steady Marangoni convection in a liquid layer heat from below and cooled from above were studied. Both viscosity variation and elasticity parameter of the surfactant act as destabilizer while the Biot number and Lewis number stabilize. For deformable surface, convection sets in at long wavelength for liquid with variable viscosity but convection occurs at short wavelength for constant viscosity liquid.

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