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Effect of interfacial deformation on the onset of convective instability in a doubly diffusive fluid layer

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Abstract—The effect of surface tension of a deformably free upper surface on the onset of convective instability in a doubly diffusive fluid layer is investigated with the linear stability theory. The eigenvalue problem is numerically solved, employing the Runge–Kutta–Gill's method of order four and making use of the Broyden's method for correction of the starting values during the integration process. The results show that the effect of Crispation number, C, becomes very sensitive on determining the possible mode of convective instability. The surface tensile and thermosolutal modes can coexist stationarily at $C = 8.44 \times 10^{-4}$. For $C < 8.16 \times 10^{-4}$, the whole system is dominated by the oscillatory mode. One of the interfacial effects, associated with the Bond number, Bo, do increase critical conditions of both stationary and oscillatory modes. The Marangoni number, M, and the solutal Marangoni number, M_S, play important roles on causing the onset of the convective instability and, in turn, reinforce each other mutually.

INTRODUCTION

For a horizontal layer of a doubly diffusive fluid with a deformably free upper surface being heated from below, the single or combined effect of the thermal or solutal buoyancy or thermally or solutally dependent surface tension may dominate the possible convection. The Rayleigh–Bénard convective instability, due to the thermal buoyancy as a result of a vertically unstable density distribution, has been extensively studied [1–4]. The Marangoni convective instability, resulting from the variation of surface tension with the temperature, can set in at the marginal state stationarily and oscillatorily [5–7]. Nield [8] analyzed the Bénard–Marangoni problem by using the linear stability analysis. The thermosolutal convective instability in a doubly diffusive fluid layer, induced by thermal and solutal gradients, was first analyzed by Nield [9]. Either thermal or solutal effect can act as a stabilizing or destabilizing factor such that stationary and oscillatory modes can be possible.

The Bénard–Marangoni convective instability, driven by thermosolutal gradients and thermosolutally dependent surface tension, has been analyzed [8–16] and has received considerable attention for its applications in engineering problems, such as oil extraction from porous media, energy storage in molten salts, crystal growth in space and colloids and detergents in chemical engineering. McTaggart [10] studied a zero-gravity environment, neglecting the effect of thermosolutal buoyancy, with a flat free upper surface under the influence of infinite surface tension. Chen and Su [11] have studied a more general case of including the effects of surface tension and interfacial deformation. Davis and Homsy [12, 13] studied the effect of the interfacial deflection, applying the energy method, and found that a deformable interface could lead to a stabilization relative to the case of a planar interface. Castillo and Velarde [14] extended the work of Davis and Homsy [13] and analyzed the role of interfacial deformation in one- and two-component fluid layer. The effect of interfacial deflection on the Marangoni instability was further studied by Scriven and Sternling [15]. Pérez-García and Carneiro [16] made a systematic study on the linear stability of the Bénard–Marangoni convective instability of single component fluid and focused on the effect of the interfacial deformation. Two stationary modes, a stationary mode and an oscillatory mode or two oscillatory modes can coexist simultaneously.

In this paper, we consider a doubly diffusive fluid layer with a deformably free upper surface under a gravity field. The surface tension is assumed to be linearly dependent on temperature and concentration and the thermosolutal buoyancy is taken into account. The principle of exchange of stabilities could hold for the convective instabilities of the Rayleigh–Bénard types [1–4] and Marangoni types [6, 7] as well. In the models considered, the free upper surface is assumed flat. Since the Crispation effect [15, 16], determining the deformability of the free upper surface, is drastic on the Marangoni convective instability, either stationary or oscillatory mode could be possible.

MATHEMATICAL FORMULATION

A horizontal fluid layer of two components with a thickness d, as shown in Fig. 1, is considered. The

NOMENCLATURE

a	wave number of small disturbance
Bo	Bond number, $\rho g d^2/\gamma$
C	Crispation number, $\mu\kappa/\gamma d$
d	thickness of fluid layer
$\mathbf{e}_z$	unit vector in the z-direction
g	gravity
h	coefficient of heat transfer
h_S	coefficient of mass transfer
k	thermal conductivity
K	parameter of solutal flux at upper surface, $h_S d/k_S$
L	parameter of heat flux at upper surface, $h\,d/k$
L_E	Lewis number, κ/κ_S
M	Marangoni number, $\tau\Delta T d/\mu\kappa$
M_S	solutal Marangoni number, $\tau_S \Delta S d/\mu\kappa$
$\mathbf{n}$	normal unit vector at free upper surface
p	pressure
Pr	Prandtl number, ν/κ
R	Rayleigh number, $\alpha g \Delta T d^3/\nu\kappa$
R_S	solutal Rayleigh number, $\alpha_S g \Delta S d^3/\nu\kappa_S$
S	solute concentration
t	time
$\mathbf{t}$	tangential unit vector at free upper surface
T	temperature
V	velocity, (u, v, w)
x, y, z	coordinates
Z	nondimensional surface deflection.

Greek symbols

α	coefficient of thermal expansion of fluid
γ	surface tension
η	position of upper free surface
θ	dimensionless temperature
κ	thermal diffusivity
μ	dynamic viscosity of fluid
ν	kinematic viscosity of fluid
ρ	density of fluid
τ	surface tension gradient with respect to temperature, $\partial\gamma/\partial T$
σ_r, σ_i	real and imaginary growth rates with time.

Superscript

$\bar{}$	steady-state quantity
$'$	dimensionless perturbed quantity.

Subscript

o	referred quantity
c	critical state
s	solutal
so	frontier point intersected by stationary and oscillatory loci.

lower boundary is rigid and isothermal and the upper boundary is deformaly free. The governing equations, assuming the Boussinessq's approximation, are

$$\rho = \rho_o[1-\alpha(T-T_o)+\alpha_S(S-S_o)] \quad (1)$$

$$\nabla \cdot V = 0 \quad (2)$$

$$\rho_o\left[\frac{\partial V}{\partial t}+(V\cdot\nabla)V\right] = -\nabla p - \rho g \mathbf{e}_z + \mu\nabla^2 V \quad (3)$$

$$\frac{\partial T}{\partial t}+(V\cdot\nabla)T = \kappa\nabla^2 T \quad (4)$$

$$\frac{\partial S}{\partial t}+(V\cdot\nabla)S = \kappa_S\nabla^2 S \quad (5)$$

where $\alpha = -\rho_o^{-1}\,\partial\rho/\partial T$ and $\alpha_S = \rho_o^{-1}\,\partial\rho/\partial S$ are the volume expansion coefficients due to variations of

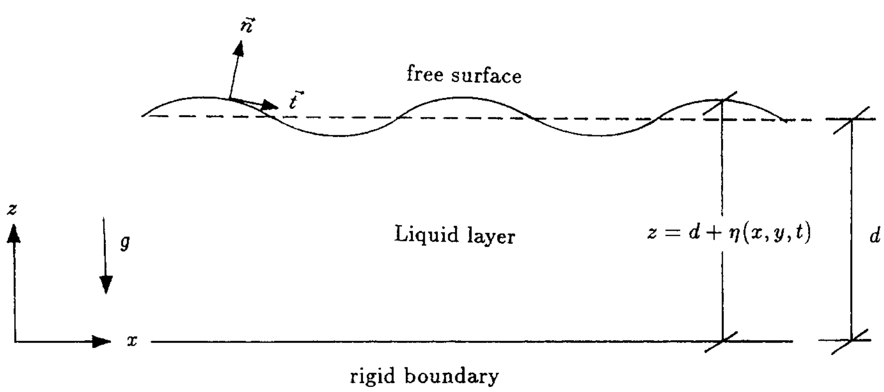

Fig. 1. Physical model.

temperature T and solute concentration S, relatively, and ρ_o, T_o and S_o are the reference density, temperature and solute concentration at the lower boundary, respectively. $V = (u, v, w)$ is the fluid velocity, p is the pressure, g is the gravity, $\mathbf{e}_z$ is the unit vector in the z-direction. The viscosity μ, the thermal diffusivity κ and the solute diffusivity κ_S are assumed constant.

The boundary conditions of the deformably free upper surface, at $z = d+\eta(x, y, t)$, are subject to momentum, energy and solute balances. The kinematic, thermal and solutal boundary conditions are

$$\frac{\partial\eta}{\partial t}+u\frac{\partial\eta}{\partial x}+v\frac{\partial\eta}{\partial y} = w \tag{6}$$

$$k\nabla T\cdot\mathbf{n}+hT = 0 \tag{7}$$

$$\kappa_S\nabla S\cdot\mathbf{n}+h_S S = 0 \tag{8}$$

where k is the thermal conductivity and h and h_S are coefficients of heat and mass transfers, respectively.

The dynamical boundary conditions of tangential and normal stresses are

$$2\mu D_{nt} = \mathbf{t}\cdot\left[\frac{\partial\gamma}{\partial T}\nabla T+\frac{\partial\gamma}{\partial S}\nabla S\right] \tag{9}$$

$$(p_a-p)+2\mu D_{nn} = \gamma\nabla\cdot\mathbf{n} \tag{10}$$

$$\gamma = \gamma_o-\tau(T-T_o)-\tau_S(S-S_o) \tag{11}$$

where p_a is the reference pressure, $\{D_{ij}\}$ is the rate of strain tensor, $\mathbf{t}$ and $\mathbf{n}$ denote the tangential and normal unit vectors at the free upper surface, γ is the surface tension, γ_o is a reference value and τ and τ_S are the rates of change of the surface tension with temperature and solute concentration, respectively.

The boundary conditions at the lower boundary of the fluid layer are rigid and isothermal with a fixed solute concentration.

The steady solutions of basic state for the temperature $\bar{T}$ and the solute concentration $\bar{S}$ are assumed linear,

$$\bar{T}(z) = T_o-\left(\frac{z}{d}\right)\Delta T \tag{12}$$

$$\bar{S}(z) = S_o-\left(\frac{z}{d}\right)\Delta S \tag{13}$$

where ΔT and ΔS are the respective differences in temperature and solute concentration across the fluid layer.

We may introduce the infinitesimal disturbances to the governing equations by setting

$$(v, u, w, \rho, p, T, S) = (0, 0, 0, \rho_o, \bar{p}, \bar{T}, \bar{S}) + (u', v', w', \rho', p', \theta', S') \tag{14}$$

where the primed quantities represent the perturbed variables.

Here the adopted scales for the coordinates, time, velocity, temperature and solute concentration are d, d^2/κ, κ/d, ΔT and ΔS, respectively. The governing equations of the perturbed state in the dimensionless form can be obtained as

$$\frac{1}{Pr}\frac{\partial}{\partial t}(\nabla^2 w') = R\nabla_h^2\theta'+\frac{R_S}{L_E}\nabla_h^2 S'+\nabla^4 w' \tag{15}$$

$$\frac{\partial\theta'}{\partial t}-w' = \nabla^2\theta' \tag{16}$$

$$\frac{\partial S'}{\partial t}-w' = \frac{1}{L_E}\nabla^2 S' \tag{17}$$

where $\nabla_h^2 = \partial^2/\partial x^2+\partial^2/\partial y^2$ and $\nabla^2 = \partial^2/\partial x^2+\partial^2/\partial y^2+\partial^2/\partial z^2$. The Prandtl number $Pr = \nu/\kappa$, the Lewis number $L_E = \kappa/\kappa_S$, and the thermal and solutal Rayleigh numbers are

$$R = \alpha g\Delta T d^3/\nu\kappa \quad R_S = \alpha_S g\Delta S d^3/\nu\kappa_S. \tag{18}$$

Similarly, the dimensionless boundary conditions of the perturbed state at the deformably free upper surface, at $z = 1$, are

$$\frac{\partial Z'}{\partial t} = w' \tag{19}$$

$$\frac{\partial\theta'}{\partial z}+L\theta' = LZ' \tag{20}$$

$$\frac{\partial S'}{\partial z}+KS' = KZ' \tag{21}$$

$$\left(\frac{\partial^2}{\partial z^2}-\nabla_h^2\right)w'+M\nabla_h^2(Z'-\theta')+\frac{M_S}{L_E}\nabla_h^2(Z'-S') = 0 \tag{22}$$

$$C\left[-\frac{1}{Pr}\frac{\partial}{\partial t}+\left(\frac{\partial^2}{\partial z^2}+3\nabla_h^2\right)\right]\frac{\partial w'}{\partial z} + (Bo-\nabla_h^2)\nabla_h^2 Z' = 0. \tag{23}$$

The Z' is the dimensionless interfacial deformation. The Crispation number C and Bond number Bo, resulting from the dynamical balance in the normal direction of the deformable surface, are defined as

$$C = \mu\kappa/\gamma d \quad Bo = \rho g d^2/\gamma. \tag{24}$$

The thermal and solutal Marangoni numbers M and M_S, being measures of the rate of change of surface tension with temperature and solute concentration, are defined as

$$M = \tau\Delta T d/\rho\nu\kappa \quad M_S = \tau_S\Delta S d/\rho\nu\kappa. \tag{25}$$

Since equations (20) and (21) are the general radiation-type conditions [9], the parameters L and K may take any value between 0 and ∞. If the upper free surface is thermally insulated, then $L = 0$. On the other hand, when $L\to\infty$, it becomes isothermal. Analoguously, if the upper free surface is impermeable, then $K = 0$. While the solute concentration at boundary is kept constant, at its saturated state, then $K\to\infty$.

The lower boundary of the fluid layer, at $z = 0$, is assumed rigid with its temperature and solute concentration fixed, thus

$$w' = \frac{\partial w'}{\partial z} = \theta' = S' = 0. \quad (26)$$

Assuming the normal mode analysis for the perturbed state, then, the perturbed vertical velocity w', temperature θ', solute concentration S' and interfacial deflection Z' become

$$(w', \theta', S', Z') = [W(z), \Theta(z), \Phi(z), Z] \times \exp[i(a_x x + a_y y) + \sigma t] \quad (27)$$

where a_x and a_y are the wave numbers of the disturbances in the x and y directions, respectively, and $\sigma = \sigma_r + i\sigma_i$ is the reaction of the disturbances to the system, σ_r is the growth rate. If $\sigma_r > 0$, the disturbances grow and the system becomes unstable. If $\sigma_r < 0$, the disturbances decay and the system becomes stable. When $\sigma_r = 0$, the instability of the system sets in at the marginal state, i.e. stationary ($\sigma_i = 0$) or oscillatory ($\sigma_i \neq 0$).

By substituting equation (27) into equations (15)–(17), the governing equations of the perturbed state become

$$\left[\frac{i\sigma_i}{Pr} - (D^2 - a^2)\right](D^2 - a^2)W = -a^2 R\Theta + a^2 \frac{R_S}{L_E}\Phi \quad (28)$$

$$[i\sigma_i - (D^2 - a^2)]\Theta - W = 0 \quad (29)$$

$$\left[i\sigma_i - \frac{1}{L_E}(D^2 - a^2)\right]\Phi - W = 0 \quad (30)$$

where the operator $D = \partial/\partial z$, and $a = (a_x^2 + a_y^2)^{1/2}$ is the wave number of the disturbances at the fluid layer.

The boundary conditions at the deformably upper free surface, at $z = 1$, are

$$W = i\sigma_i Z \quad (31)$$

$$(D + L)\Theta = LZ \quad (32)$$

$$(D + K)\Phi = KZ \quad (33)$$

$$(D^2 + a^2)W + Ma^2(\Theta - Z) + \frac{M_S}{L_E}a^2(\Phi - Z) = 0 \quad (34)$$

$$C\left[\frac{i\sigma_i}{Pr} - (D^2 - 3a^2)\right]DW + (Bo + a^2)a^2 Z = 0 \quad (35)$$

and the boundary conditions at the bottom surface, at $z = 0$, is

$$W = DW = \Theta = \Phi = 0. \quad (36)$$

NUMERICAL PROCEDURE

The governing equations (28)–(30) and boundary conditions (31)–(36) form a Sturm–Liouville's problem with the thermal or solutal Marangoni number, M or M_S, or thermal or solutal Rayleigh number, R or R_S, being the eigenvalue with other physical parameters, such as Pr, L_E, C, Bo, L, K and a fixed. The shooting technique, based on the Runge–Kutta–Gill's method of order four [17], is used to solve the problem.

The first step in the procedure is to rewrite equations (28)–(30) as a system of first-order equations,

$$W = u_1$$

$$DW = Du_1 = u_2 \quad (37)$$

$$D^2 W = Du_2 = u_3 \quad (38)$$

$$D^3 W = Du_3 = u_4 \quad (39)$$

$$\Theta = u_5$$

$$D\Theta = Du_5 = u_6 \quad (40)$$

$$\Phi = u_7$$

$$D\Phi = Du_7 = u_8 \quad (41)$$

and we obtain

$$D^4 W = Du_4 = \left(\frac{i\sigma_i}{Pr} + 2a^2\right)u_3 - \left(\frac{i\sigma_i}{Pr} + a^2\right)a^2 u_1 + Ra^2 u_5 - \frac{R_S}{L_E}a^2 u_7 \quad (42)$$

$$D^2\Theta = Du_6 = (i\sigma_i + a^2)u_5 - u_1 \quad (43)$$

$$D^2\Phi = Du_8 = (L_E i\sigma_i + a^2)u_7 - L_E u_1. \quad (44)$$

The shooting procedure starts from the upper boundary conditions (31)–(34), at $z = 1$, and trys to match the lower boundary conditions (36), at $z = 0$.

The upper boundary conditions (31)–(34), at $z = 1$, can be expressed as

$$u_1 = \left[-\frac{3i\sigma_i C}{Bo + a^2} + \frac{\sigma_i^2 C}{(Bo + a^2)a^2 Pr}\right]u_2 + \frac{i\sigma_i C}{Boa^2 + a^4}u_4 \quad (45)$$

$$u_3 = \left[\frac{3i\sigma_i Ca^2}{Bo + a^2} - \frac{\sigma_i^2 C}{(Bo + a^2)Pr} - \frac{3MCa^2}{Bo + a^2} - \frac{i\sigma_i MC}{(Bo + a^2)Pr} - \frac{3M_S Ca^2}{(Bo + a^2)L_E} - \frac{i\sigma_i M_S C}{(Bo + a^2)L_E Pr}\right]u_2 - \left[\frac{i\sigma C}{Bo + a^2} - \frac{MC}{Bo + a^2} - \frac{M_S C}{(Bo + a^2)L_E}\right]u_4 - Ma^2 u_5 - \frac{M_S}{L_E}a^2 u_7 \quad (46)$$

$$u_6 = -\left[\frac{3LC}{Bo + a^2} + \frac{i\sigma_i LC}{(Bo + a^2)a^2 Pr}\right]u_2 + \frac{LC}{Bo + a^2}a^2 u_4 - Lu_5 \quad (47)$$

$$u_8 = -\left[\frac{3KC}{Bo+a^2} + \frac{i\sigma_i KC}{(Bo+a^2)a^2 Pr}\right]u_2 + \frac{KC}{Bo+a^2}a^2 u_4 - Ku_7. \quad (48)$$

We shall guess four boundary conditions,

$$u_2 = c_1, \quad u_4 = c_2, \quad u_5 = c_3, \quad \text{and} \quad u_7 = c_4 \quad (49)$$

then the general form of the solution becomes

$$\underline{U} = c_1 U_1 + c_2 U_2 + c_3 U_3 + c_4 U_4 \quad (50)$$

where

$$\underline{U} = [u_1, u_2, u_3, u_4, u_5, u_6, u_7, u_8]^{\mathrm{T}} \quad (51)$$

$$U_1 = \left[-\frac{3i\sigma_i C}{Bo+a^2} - \frac{\sigma_i^2 C}{(Bo+a^2)a^2 Pr},\ 1,\ \frac{3i\sigma_i Ca^2}{Bo+a^2} - \frac{\sigma_i^2 C}{(Bo+a^2)Pr} - \frac{3MCa^2}{Bo+a^2} - \frac{i\sigma_i MC}{(Bo+a^2)Pr} - \frac{3M_S Ca^2}{(Bo+a^2)L_E} - \frac{i\sigma_i M_S C}{(Bo+a^2)L_E Pr},\ 0,\ 0,\ -\frac{3LC}{Bo+a^2} - \frac{i\sigma_i LC}{(Bo+a^2)a^2 Pr}, 0,\ -\frac{3KC}{Bo+a^2} - \frac{i\sigma_i KC}{(Bo+a^2)a^2 Pr}\right]^{\mathrm{T}} \quad (52)$$

$$U_2 = \left[\frac{i\sigma_i C}{Boa^2+a^4},\ 0,\ -\frac{i\sigma C}{Bo+a^2} + \frac{MC}{Bo+a^2} + \frac{M_S C}{(Bo+a^2)L_E},\ 1,\ 0,\ \frac{LC}{Bo+a^2}a^2,\ 0,\ \frac{KC}{Bo+a^2}a^2\right]^{\mathrm{T}} \quad (53)$$

$$U_3 = [0, 0, -Ma^2, 0, 1, -L, 0, 0]^{\mathrm{T}} \quad (54)$$

$$U_4 = \left[0, 0, -\frac{M_S}{L_E}a^2, 0, 0, 0, 1, -K\right]^{\mathrm{T}}. \quad (55)$$

Guess a value for M and assume U_i, $i = 1, 4$, in (52)–(55) as a set of initial conditions. We then start the shooting procedure, using the Runge–Kutta–Gill's method of order four, from $z = 1$ and try to match the lower boundary conditions at $z = 0$. The results finally turn into a matrix form,

$$\begin{bmatrix} U_1^1 & U_2^1 & U_3^1 & U_4^1 \\ U_1^2 & U_2^2 & U_3^2 & U_4^2 \\ U_1^5 & U_2^5 & U_3^5 & U_4^5 \\ U_1^7 & U_2^7 & U_3^7 & U_4^7 \end{bmatrix} \begin{bmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \end{bmatrix} = 0 \quad (56)$$

where the superscript indicates the element of U_i, $i = 1, 4$. The determinant of the matrix of equation (56) is complex and both real and imaginary parts should vanish individually for c_i being nontrivial such that the relation among the parameters is thus established,

$$f(i\sigma_i, R, R_S, M, M_S, C, Bo, K, L, Pr, L_E, a) = 0 \quad (57)$$

where f is the determinant of the coefficient matrix.

With fixed values for the physical parameters R, R_S, M_S, C, Bo, K, L, Pr, L_E and the wave number a, the Marangoni number M and frequency σ_i, using the iterative Broyden's method [18], is solved. The critical conditions M_c and σ_{ic}, marking the onset of convective instability at the marginal state, are thus obtained.

RESULTS AND DISCUSSION

A. *Stationary convection*

The numerical results are checked and compared with the previous works. McTaggart[10] studied the case that the surface tension is thermally and solutally dependent and the upper surface is flatly free (i.e. the Crispation number $C = 0$). In absence of the thermosolutal convection (i.e. $R = 0$ and $R_S = 0$), the critical conditions M_c and a_c of the stationary modes are shown in Table 1 for $C = 0$, and in Table 2 for $C \neq 0$. The results, obtained by Pérez-García and Carneiro [16] in without the solutal effects (i.e. $M_S = 0$), are also confirmed. The effects of interfacial deformation, depending on Crispation number C, Bond number Bo and the degree of conductivity and permeability at the upper boundary (i.e. K and L) do have great influences on the onset of convective instabilities. Here, we take $K = L = 0$ for analysis in this paper.

For Marangoni convective instability only (i.e. $R = 0$ and $R_S = 0$), Fig. 2 presents the critical Marangoni number M_c as a function of the Crispation number C for selected values of the solutal Marangoni number $M_S = -100$, 0 and 100. A negative value of M_S, related to an increasing surface tension with an increasing solute concentration, would suppress the whole system to a more stabilizing state. The Crispation number, associated with the inverse effect of the surface tension, shows the rigidity of the free upper surface. For the Crispation number C to be zero, the free upper surface, subject to an infinite surface tension, is taken to be undeformably flat and the system becomes more stabilizing for both Marangoni and thermosolutal convective instabilities. For C to be very large, the free upper surface, subject to a vanishing surface tension, becomes deformable and the system tends to be less stabilizing for both Marangoni and thermosolutal convective instabilities. The critical Marangoni number M_c decreases as C increases. These decreasing trends are negligible for $0 \leqslant C \leqslant 10^{-4}$, in which range the critical wave numbers, associated with the thermosolutal modes, are finite. The decreasing trends become proportional and significant for $C \geqslant 10^{-3}$, in which range the criti-

Table 1. Critical values of Marangoni number M_c and the corresponding critical wave number a_c for different values of L and K on the stationary stability of convection without the thermosolutal convection ($C = 0$, $Bo = 0.1$, $R = R_S = 0$ and $L_E = 1$)

		$M_S = -50$		$M_S = 0$		$M_S = 100$	
		[10]	This study	[10]	This study	[10]	This study
		M_c	M_c	M_c	M_c	M_c	M_c
L	K	a_c	a_c	a_c	a_c	a_c	a_c
0	0	129.6	129.603	79.6	79.603	−20.4	−20.396
		1.99	1.993	1.99	1.993	1.99	1.993
0	1	113.1	113.063	79.6	79.603	11.2	11.207
		1.89	1.891	1.99	1.993	2.21	2.221
1	0	187.2	187.231	116.1	116.121	−30.4	−30.391
		2.40	2.401	2.25	2.246	1.93	1.930
1	1	166.1	166.121	116.1	116.121	16.1	16.121
		2.25	2.246	2.25	2.246	2.25	2.246
5	5	300.6	300.580	250.6	250.579	150.6	150.579
		2.60	2.598	2.60	2.598	2.60	2.598
10	10	463.4	463.408	413.4	413.408	313.4	313.408
		2.74	2.743	2.74	2.743	2.74	2.743
10^{10}	10^{10}	3.2×10^{11}	3.2×10^{11}	3.2×10^{11}	3.2×10^{11}	3.2×10^{11}	3.2×10^{11}
		3.01	3.009	3.01	3.009	3.01	3.009
10^{10}	0	4.7×10^{11}	4.7×10^{11}	3.2×10^{11}	3.2×10^{11}	-1.0×10^{11}	-1.0×10^{11}
		3.66	3.660	3.01	3.009	1.82	1.819
10^{10}	10^{10}	79.6	79.603	79.6	79.603	79.6	79.603
		1.99	1.993	1.99	1.993	1.99	1.993

Table 2. Critical values of Marangoni number M_c and the corresponding critical wave number a_c for different values of C and M_S on the stationary stability of convection without the thermosolutal convection ($Bo = 0.1$, $R = R_S = 0$ and $L_E = 1$)

	$M_S = -100$		$M_S = 0$				$M_S = 100$	
	This study		Data from [16]		This study		This study	
C	M_c	a_c	M_c	a_c	M_c	a_c	M_c	a_c
0	179.603	1.993	79.607	1.99	79.603	1.993	−20.396	1.993
10^{-6}	179.602	1.993	79.606	1.99	79.602	1.993	−20.397	1.993
10^{-5}	179.592	1.993	79.596	1.99	79.592	1.993	−20.408	1.993
10^{-4}	179.496	1.989	79.499	1.99	79.496	1.989	−20.496	1.989
10^{-3}	166.667	0.001	66.667	0.00	66.667	0.001	−33.333	0.001
10^{-2}	106.667	0.001	6.667	0.00	6.667	0.001	−93.333	0.001
10^{-1}	100.667	0.001	0.667	0.00	0.667	0.001	−99.333	0.001

cal wave numbers, related to the surface tensile modes, are vanishing.

There are, on the neutral curve, two minimal points, corresponding to the thermosolutal mode of finite wave number and the surface tensile mode of small wave number respectively and the mode with the smaller value of Marangoni number becomes dominant. In particular, when the two minimal points have the same critical Marangoni number, both modes would set in simultaneously. Therefore, there exists a frontier point in the range $[2 \times 10^{-4}, 5 \times 10^{-3}]$ of the Crispation number C at which both modes coexist simultaneously and across which a jump on the convective instability from one mode of finite critical wave number to another mode of a vanishing one does exist. Figure 2 shows that the critical Marangoni number M_c at the frontier points as marked occurs at $C = 8.44 \times 10^{-4}$ with its values being 179.365, 79.365, −20.634 for $M_S = -100$, 0, 100, respectively. In absence of thermosolutal buoyancy, the Crispation number C at the frontier points seems to be invariant to the solutal Marangoni number M_S.

The Bond number Bo, being a ratio of the relative effect of gravity to surface tension, measures the dominant effect of the two in flattening the free upper surface. The Marangoni number M as a function of the wave number a is shown in Fig. 3 for various values of the Bond number Bo. The first minimal point, associated with the surface tensile modes of zero wave number, are very sensitive to the surface tension and increase as Bo increases. But no such trend is shown to the second minimal point, associated with the thermosolutal modes of finite wave number. Figure 4 shows the critical Marangoni number M_c as a function of Bo for selected values of $M_S = -100$, 0 and 100. The critical Marangoni number M_c varies linearly with Bo, for $Bo < 0.1$ in which range the surface tensile mode prevails. M_c varies slightly with Bo,

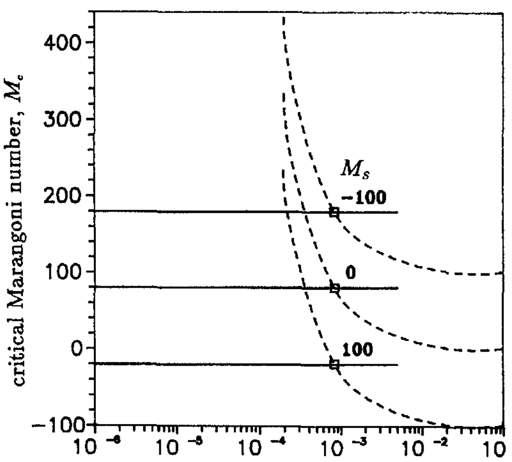

Fig. 2. The effect of C and M_S on the stationary stability of convection without the thermosolutal convection, $Bo = 0.1$, $R = R_S = K = L = 0$; □, $C = 8.44 \times 10^{-4}$; dashed line, the surface tensile mode; solid line, the thermosolutal mode.

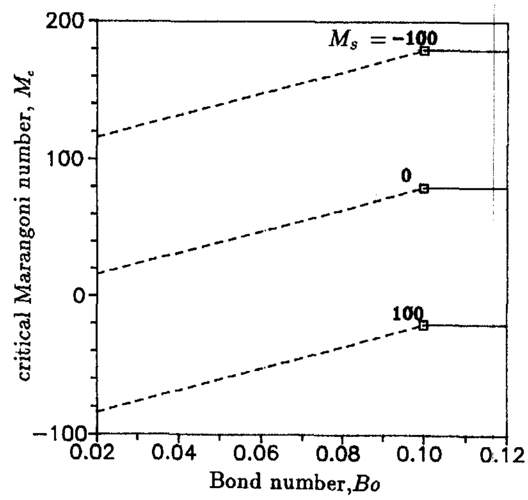

Fig. 4. The effect of Bo and M_S on the stationary stability of convection for $C = 8.44 \times 10^{-4}$, $R = R_S = K = L = 0$, $L_E = 1$; □, $Bo = 0.1$; dashed line, the surface tensile mode; solid line, the thermosolutal mode.

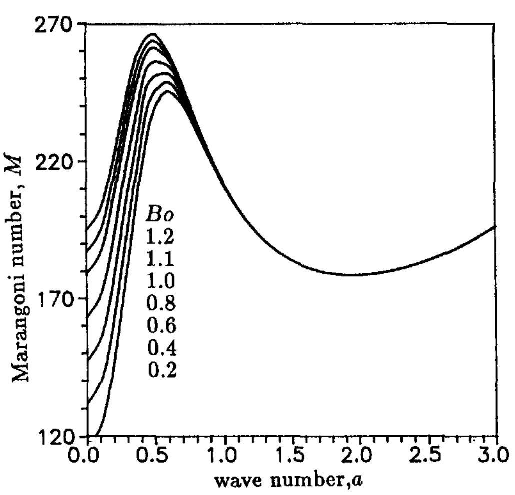

Fig. 3. The stationary neutral curves $M(a)$ are plotted for several values of Bo on the stationary stability convection for $M_S = -100$, $R = R_S = K = L = 0$, $L_E = 1$ and $C = 8.44 \times 10^{-4}$.

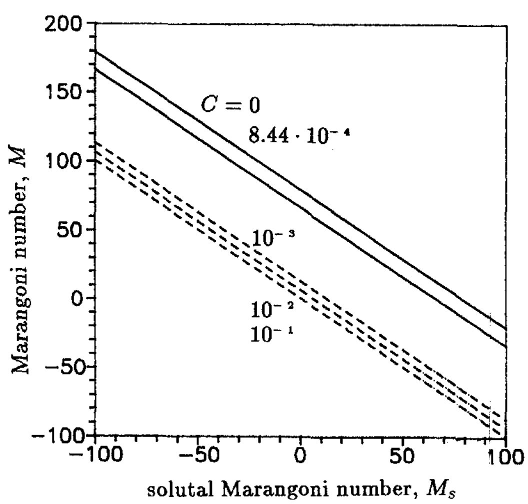

Fig. 5. Locus of Marangoni number M and solutal Marangoni number M_S for different values of C on the stationary stability of convection for $Bo = 0.1$, $R = R_S = K = L = 0$, $L_E = 1$; dashed line, the surface tensile mode; solid line, the thermosolutal mode.

for $Bo > 0.1$, in which range the thermosolutal mode dominates. As Bo increases up to a frontier point at 0.1, at which these two modes can coexist simultaneously, as shown in Fig. 4 for $C = 8.44 \times 10^{-4}$, and across which convective instabilities switch from one mode to another. However, without the effect of thermosolutal buoyancy, the Bond number Bo at the frontier point is invariant to the solutal Marangoni number M_S. The critical values in the plane (M, M_S) is plotted in Fig. 5 for selected values of the Crispation number $C = 0$, 8.44×10^{-4}, 10^{-3}, 10^{-2} and 10^{-1}. It is shown that M decreases with M_S linearly. The two agencies, causing the mechanism of convective instabilities, would reinfore each other and the stability curve in the (M, M_S)-plane do satisfy the following linear relation [10]

$$M = M_c - M_S \tag{58}$$

where M_c, depending on the value of the Crispation number, are obtained from Table 2 for $M_S = 0$.

The influence of the conductivity and permeability of the free upper surface can be appreciated from Fig. 6 for thermosolutal mode ($C \leqslant 8.44 \times 10^{-4}$) and surface tensile mode ($C = 10^{-1}$), as the thermosolutal buoyancy is being neglected (i.e. $R = R_S = 0$). Three types of boundary conditions are considered: (a) conductive and impermeable ($L = 1$ and $K = 0$), (b) insulating and impermeable ($L = K = 0$) and (c) insulating and permeable ($L = 0$ and $K = 1$). It is shown

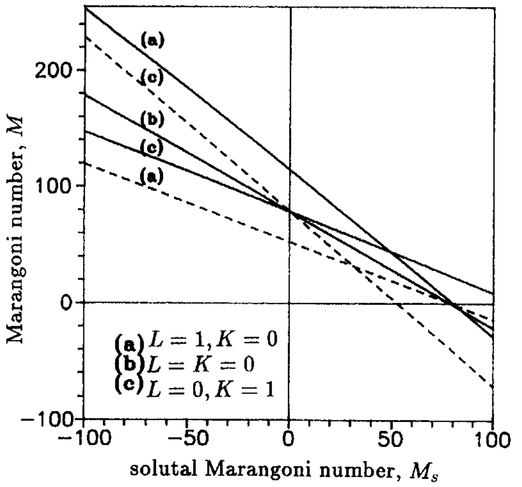

Fig. 6. Locus of Marangoni number M and solutal Marangoni number M_S for different values of L and K on the stationary stability of convection for $Bo = 0.1$, $R = R_S = 0$, $L_E = 1$; dashed line, $C = 10^{-1}$; solid line, $C = 8.44 \times 10^{-4}$.

from Fig. 6 that the conductivity and permeability tend to have contrary effects on surface tensile modes with respective to thermosolutal mode. For thermosolutal mode with the negative solutal Marangoni number M_S, the curve of case (a) with $L = 1$ and $K = 0$ lies above that of case (b) with $L = K = 0$, which, in turn, lies above that of case (c) with $L = 0$ and $K = 1$. However, this trend turns inverse for the positive solutal Marangoni number M_S.

As we know that the thermosolutal buoyancy acts as a main mechanism of driving convective instabilities. The neutral curves of Marangoni number M as a function of wave number a are ploted in Fig. 7 for various Rayleigh number R and $C = 8.44 \times 10^{-4}$. As expected, two different kinds of instabilities are shown: thermosolutal mode and surface tensile modes. All of the curves of the surface tensile modes do coincide and reach to the same minimum value in the limit $a \to 0$. The second minimal point of the thermosolutal mode descends, as the Rayleigh number R increases.

The critical Marangoni number M_c as a function of the Rayleigh number R is plotted in Fig. 8 for several values of the Crispation number $C = 8.44 \times 10^{-4}$, 10^{-3}, 2.5×10^{-3}, 5×10^{-3} and the solutal Marangoni number $M_S = -100$. We obtain a curve similar to that achieved by Pérez-García and Carneiro [16] without the solutal effect. When $C \leqslant 8.44 \times 10^{-4}$, the thermosolutal mode dominates. For a fixed value of the Crispation number C, for example 10^{-3}, as the Rayleigh number R increases, the surface tensile mode dominates up to a frontier point of $R = 107$, after which the thermosolutal mode prevails. The Rayleigh number R at the frontier point increases as C increases and the values are 107, 473 and 588 for $C = 10^{-3}$, 2.5×10^{-3} and 5×10^{-3}, respectively.

The solutal effects do play an important role for the onset of the convective instabilities, depending on the positive or negative values of the solutal Rayleigh number R_S and Marangoni number M_S. A positive value of R_S means an increasing distribution of the light component with height and the system tends to be more stabilizing. The effects of M_S are similar to that of M. A negative M_S would increase the effect of the surface tension and, thus, give rise to a larger critical value. From Fig. 9, with $C = 8.44 \times 10^{-4}$, the second minimal point of the thermosolutal mode increases by increasing the solutal Rayleigh number R_S.

All of the curves of the surface tensile modes, as discussed before, do coincide and reach to the same minimum value in the limit $a \to 0$. The critical Mar-

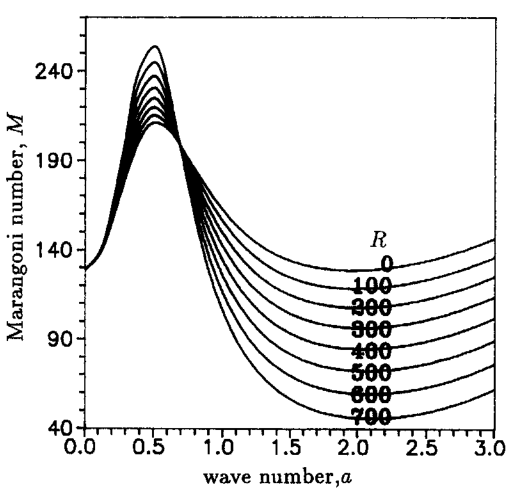

Fig. 7. The stationary neutral curves $M(a)$ are plotted for several values of R on the stationary stability convection for $M_S = -100$, $R_S = K = L = 0$, $L_E = 1$ and $C = 8.44 \times 10^{-4}$.

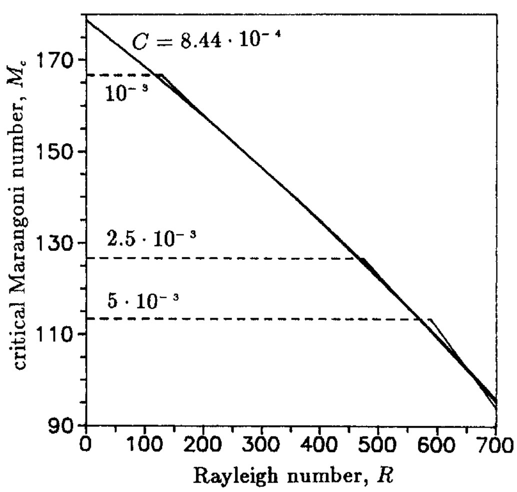

Fig. 8. Locus of Marangoni number M and Rayleigh number R for different values of C on the stationary stability of convection for $Bo = 0.1$, $M_S = -100$, $R_S = K = L = 0$, $L_E = 1$; dashed line, the surface tensile mode; solid line, the thermosolutal mode.

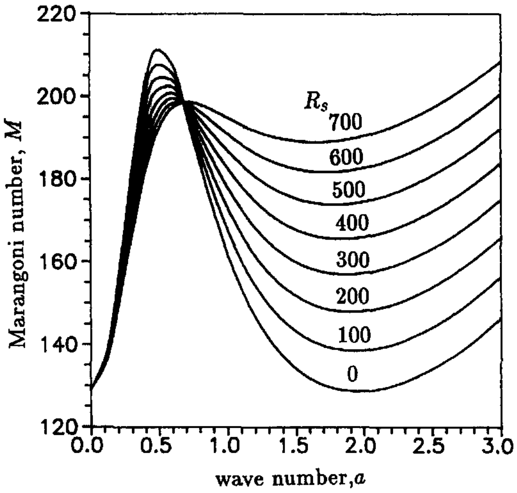

Fig. 9. The stationary neutral curves $M(a)$ are plotted for several values of R_S on the stationary stability convection for $M_S = -100$, $R = K = L = 0$, $L_E = 1$ and $C = 8.44 \times 10^{-4}$.

angoni number M_c as a function of the solutal Rayleigh number R_S is plotted in Fig. 10 for C being 10^{-3}, 8.44×10^{-4}, 7×10^{-4}, 6×10^{-4}, 5×10^{-4} and 10^{-4}. When $C \geqslant 8.44 \times 10^{-4}$, the surface tensile modes (dashed lines) dominate. But, when $C < 8.44 \times 10^{-4}$, the surface tensile modes (dashed lines) with zero wave number start to appear and compete with the thermosolutal modes for dominating the onset of the system. The mode with an absolute minimun value of Marangoni number has a critical one and would set in at the marginal state. As the solutal Rayleigh number R_S increases from zero, the thermosolutal mode of the system is more stabilizing and the surface tensile mode becomes dominant. At the frontier point, where thermosolutal and surface tensile modes coexist simultaneously, the solutal Rayleigh number R_S being 161, 325 and 564 for $C = 7 \times 10^{-4}$, 6×10^{-4} and 5×10^{-4}, respectively, do increase as C decreases.

B. *Oscillatory convection*

The behavior of convective instability in a horizontal layer of a two-component fluid with a deformably free upper surface is not restrained to the stationary convection only [12, 16]. McTaggart [10] had studied the oscillatory instability of convection, driven by concentration and temperature-dependent surface tension, with the upper surface flat and free. In absence of the thermosolutal buoyancy and for an infinite surface tension (i.e. $R = R_S = 0$ and $C = 0$), we have the critical Marangoni number $M_c = 85.199$ and the frequency $\sigma_{ic} = -0.720$ which concide exactly with McTaggart [10] for $Pr = 2$ and $M_S = -50$. Figures 11(a) and (b) present marginal curves of the oscillatory modes of the Marangoni number M and frequency σ_i vs the wave number a for selected values of C. The effect of C is very similar to both stationary and oscillatory modes. As C increases, the Marangoni number M of surface tensile modes are reduced. Those decreasing trends are negligible for $0 \leqslant C \leqslant 10^{-4}$, to which range the thermosolutal mode dominates, and become proportionally significant for $C \geqslant 10^{-3}$, to which range the surface tensile mode prevails. For a possible case of the latter, the corresponding critical wave number is not necessarily vanishing. As the Crispation number C is approximately equal to 10^{-3}, the neutral curve presents two minimal points, indicating the existence of the thermosolutal and surface tensile modes. The critical Marangoni number M_c and its related frequency σ_{ic} for both stationary and oscillatory modes are listed and compared in Table 3. There always exists a Crispation number C_o^* such that for $C > C_o^*$ the minimal Marangoni number for the stationary modes is smaller than that for the oscil-

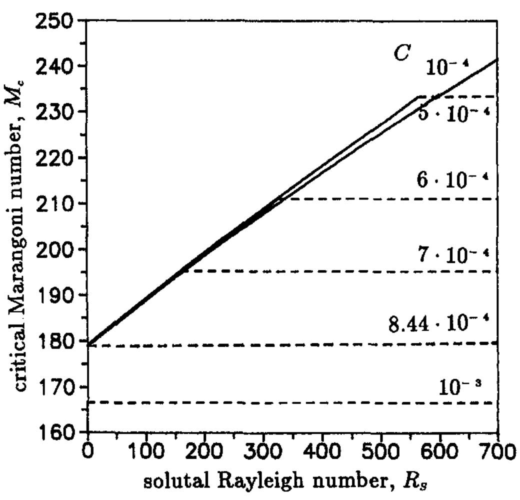

Fig. 10. Locus of Marangoni number M and solutal Rayleigh number R_S for different values of C on the stationary stability of convection for $Bo = 0.1$, $M_S = -100$, $R = K = L = 0$, $L_E = 1$; dashed line, the surface tensile mode; solid line, the thermosolutal mode.

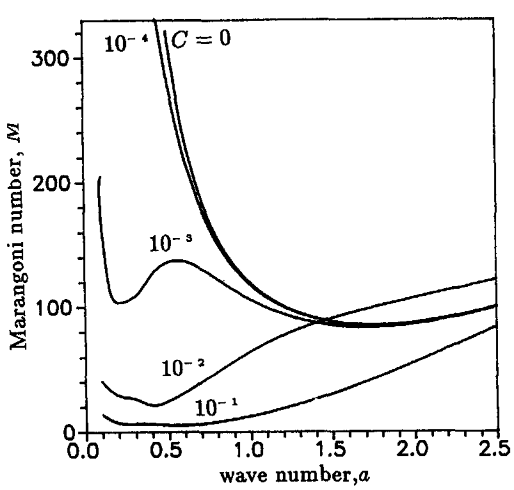

Fig. 11. (a) The oscillatory neutral curves $M(a)$ are plotted for several values of C on the stability of convection for $Pr = 2$, $M_S = -50$, $L_E = 25$, $R = R_S = K = L = 0$, $Bo = 0.1$.

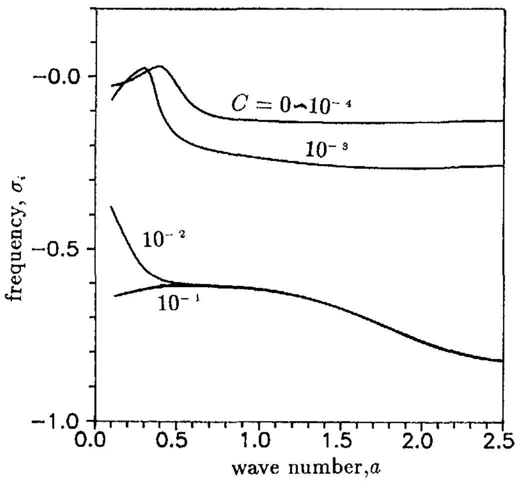

Fig. 11. (b) The oscillatory frequency neutral curves $\sigma_i(a)$ are plotted for several values of C on the stability of convection for $Pr = 2$, $M_S = -50$, $L_E = 25$, $R = R_S = K = L = 0$, $Bo = 0.1$.

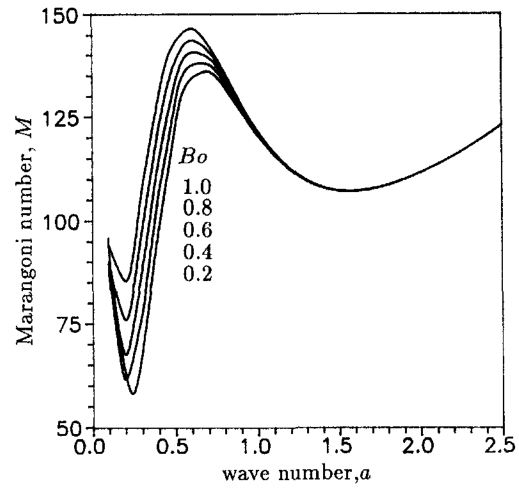

Fig. 12. (a) The oscillatory neutral curves $M(a)$ are plotted for several values of Bo on the stability of convection for $C = 10^{-3}$, $Pr = 2$, $M_S = -50$, $L_E = 25$, $R = R_S = K = L = 0$.

latory ones. The convective instability takes place stationarily in the form of the surface tensile modes. It is shown numerically that $C_o^* = 8.16 \times 10^{-4}$. The physical behavior for $C < C_o^*$ is contrary to that for $C > C_o^*$. The minimal Marangoni number for the oscillatory modes is smaller than that for the stationary ones. The convective instability takes place oscillatorily in the form of the thermosolutal modes.

The Bond number Bo, associated with the ratio of gravity to surface tension, measures the dominant effect in flattening a curved free surface. As shown in Figs. 12(a) and (b), the effects of Bo to stationary and oscillatory modes are quite similar also. The minimal Marangoni number M and its related frequency σ_i of the surface tensile modes with a smaller but not vanishing wave number increases as Bo increases. It is worth noting that from Table 3 for $C = 10^{-3}$ the minimal Marangoni number M and its corresponding wave number a for the oscillatory modes are greater than those for the stationary ones.

The critical Marangoni number M_c as a function of

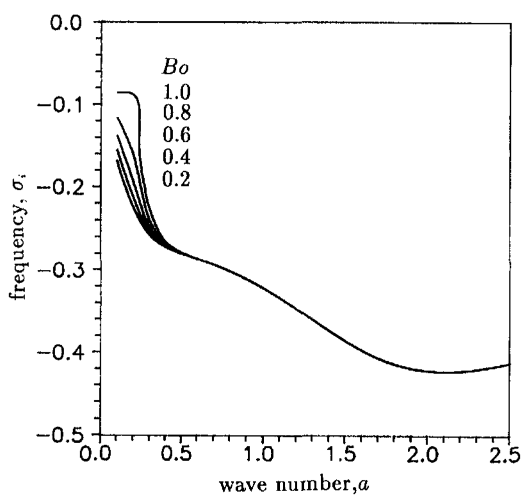

Fig. 12. (b) The oscillatory frequency curves $\sigma_i(a)$ are plotted for several values of Bo on the stability of convection for $C = 10^{-3}$, $Pr = 2$, $M_S = -50$, $L_E = 25$, $R = R_S = K = L = 0$.

Table 3. Critical values of Marangoni number M_c and the corresponding critical wave number a_c for different values of C on the stationary and oscillatory stability of convection without the thermosolutal convection ($M_S = -50$, $Pr = 2$, $Bo = 0.1$, $R = R_S = 0$ and $L_E = 25$)

C	Stationary			Oscillatory		
	M_c	a_c	σ_{ic}	M_c	a_c	σ_{ic}
0	129.603	1.993	0	85.199	1.759	−0.720
10^{-6}	129.602	1.993	0	85.198	1.759	−0.720
10^{-5}	129.587	1.992	0	85.182	1.759	−0.719
10^{-4}	129.431	1.986	0	85.028	1.713	−0.719
8.16×10^{-4}	83.701	1.934	0	83.776	1.722	−0.710
10^{-3}	68.667	0.001	0	83.447	1.713	−0.703
10^{-2}	8.667	0.001	0	21.099	0.403	−0.100
10^{-1}	2.667	0.001	0	5.026	0.550	−0.059

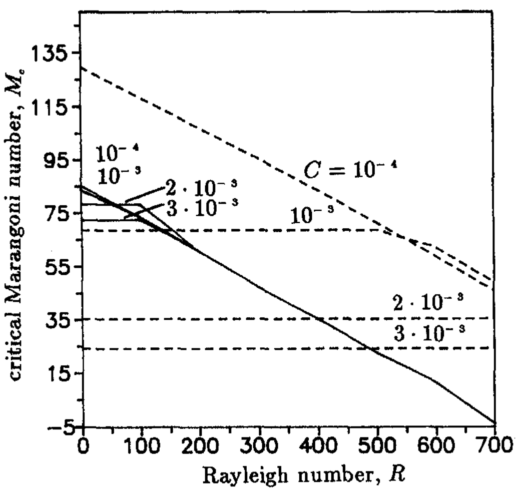

Fig. 13. Locus of Marangoni number M and Rayleigh number R for different values of C on the instability of convection for $Pr = 2$, $M_S = -50$, $L_E = 25$, $R_S = K = L = 0$, $Bo = 0.1$; dashed line, the stationary mode; solid line, the oscillatory mode.

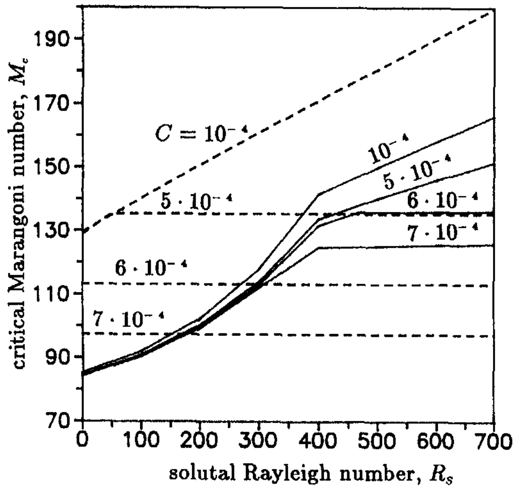

Fig. 14. Locus of Marangoni number M and solutal Rayleigh number R_S for different values of C on the instability of convection for $Pr = 2$, $M_S = -50$, $L_E = 25$, $R = K = L = 0$, $Bo = 0.1$; dashed line, the stationary mode; solid line, the oscillatory mode.

the Rayleigh number R, in absence of solutal buoyancy, are plotted in Fig. 13 for both stationary and oscillatory modes at several selected values of the Crispation number C. The dashed lines correspond to the stationary modes, while the solid lines correspond to the oscillatory modes. The effect of C on the stationary surface tensile modes is more significant than that on the oscillatory ones, but this effect becomes negligible to both stationary and oscillatory thermosolutal modes. However, the surface tensile modes, stationary or oscillatory, are very indifferent to the thermal Rayleigh number R, especially for $C > 10^{-3}$. While, both stationary and oscillatory thermosolutal modes depends strongly on the thermal Rayleigh number R. The Critical Marangoni number M_c of the oscillatory thermosolutal modes decrease as the thermal Rayleigh number R increases. For the instability of the system under consideration, there exists possibly three types of frontier points, intersected by two stationary, two oscillatory and one stationary and one oscillatory loci. For a fixed value of Crispation number C, the minimal Marangoni number of the frontier points of the former two types, being smaller than those of stationary surface tensile modes or oscillatory thermosolutal modes, are theoretically interesting, but not important at the critical state. The point, intersected by stationary and oscillatory loci as shown in Fig. 13, represent the frontier one, at which both modes do set in simultaneously. The frontier points, for $C = C_{so}$ being 10^{-3}, 2×10^{-3} and 3×10^{-3}, occur when the thermal Rayleigh number $R = R_{so}$ are 142, 419 and 508, respectively. The frontier points would vanish for $C > 10^{-4}$, in which range the surface tension would suppress the surface tensile modes, stationary or oscillatory, and the minimal Marangoni numbers of thermosolutal modes, stationary and oscillatory, decrease linearly with the Rayleigh number R with the oscillatory one being dominant. For $C \geqslant 10^{-3}$, the stationary surface tensile modes, having a smaller minimal Marangoni number, dominate for R being less than R_{so}, while the oscillatory thermosolutal modes become prevailing for R being greater than R_{so}.

Loci of critical Marangoni number M_c and solutal Rayleigh number R_S of stationary and oscillatory modes, in absence of the thermal buoyancy, are plotted in Fig. 14 at several selected values of the Crispation number C. The positive solutal Rayleigh number corresponds to an increasing light component with height and, as well, a more stabilizing density distribution, which illustrates the contrast result caused by the positive Rayleigh number R. While the negative solutal Marangoni number M_S gives rise to an increasing surface tension with the solute concentration. The effect of C is, as before, more decisive on the stationary modes than on the oscillatory ones. The critical Marangoni number M_c of the stationary thermosolutal modes increases linearly with the solutal Rayleigh number R_S, for $C \leqslant 10^{-4}$, and that of the stationary thermosolutal modes becomes unaltered, for $C > 10^{-4}$. The oscillatory modes, depending strongly on C and R_S, behave differently to the stationary modes. The critical Marangoni number M_c increase with the solutal Rayleigh number R_S largely for the thermosolutal modes and slowly, not indifferently instead, for the surface tensile modes. For the thermosolutal modes, variation of M_c is negligibly small, especially at small values of the solutal Rayleigh number for the different values of Crispation number. The competition between the stationary and oscillatory modes determines the favorite mode, prevailing in the system. The frontier points, for $C = C_{so}$ being 5×10^{-4}, 6×10^{-4} and 7×10^{-4}, occur when the solutal Rayleigh number R_{Sso} are 427, 309 and 159, respectively, and both modes prevail simultaneously.

CONCLUSIONS

The effect of interfacial deformation on the onset of convective instability in a doubly diffusive fluid layer has been studied theoretically and numerically. The following results are obtained.

(1) There exists stationary or oscillatory convective instabilities of thermal or solutal modes of buoyancy or surface tension dependence with finite wave numbers and of surface tensile modes with vanishing ones.

(2) For stationary instabilities, both surface tensile and thermosolutal modes can co-exist at the Crispation number $C = 8.44 \times 10^{-4}$. For $C < 8.16 \times 10^{-4}$, the system sets in oscillatorily.

(3) The thermal gradient is a destabilizing factor and, as well, the solutal gradient is a stabilizing one. Both effects become important for the thermosolutal modes.

(4) The Bond number Bo, sensitive to the surface tensile modes, increases the critical conditions of both stationary and oscillatory modes.

(5) The Marangoni convection, driven by the surface tension, is strongly subject to the thermal and solutal gradients and the thermal and solutal Marangoni numbers. The thermal and solutal effects, being the main mechanisms of causing the convective instabilities of the system, may reinforce each other mutually. The stationary locus between thermal and solutal Marangoni numbers satisfies the linear relation.

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